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COMPUTER SIMULATION OF THE CONTINUOUS THE PROCESS. VOLUME 1: THE NITRATION SECTION

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January 1975

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### SUMMARY

The development of a mathematical model of the nitration section of a continuous process for the manufacture of TNT is described. Implementation of the model on a high-speed digital computer is also discussed, as are the steady state and dynamic studies that were conducted on the resulting computer simulation. The way in which optimized process operating conditions and improved process control strategies were developed using the simulation is illustrated.

#### INTRODUCTION

Computer simulation of a chemical process, simply defined, is a procedure by which a mathematical model of the process' relevant chemical and physical characteristics is first constructed, after which the resultant system of equations is programmed for solution on a high speed computer. Such a model is usually based upon a combination of prevailing theory and empirical relationships, and, when properly formulated, will predict all process input/output relationships. The applicability of such a model depends heavily upon the ratio of theory to empiricism employed in model development. For example, a totally empirical model such as a least squares polynomial approximation is useful only within the boundaries of experimental observation, whereas a totally theoretical model, for example Newton's Laws of Motion as a description of projectile trajectories, will apply under virtually all conditions. For most complex systems (such as a chemical process) an optimum balance between theory and observation must be sought.

Upon completion of mathematical modeling and computer programming, the resulting computer simulation can then, after verification against real process data, be used to study the process without incurring the cost, time loss, and (especially in the case of explosives manufacturing processes) the risk involved in pilot plant or full-scale plant experimentation. Most often, studies of both the static and dynamic states of the process are carried out, the objective of the former being optimization of operating and design conditions, and the objective of the latter being development and testing of control strategies.

It has always been possible to develop mathematical models of complex physical systems. However, the value of such models remained questionable until the application of digital, analog, and hybrid computers made possible the rapid solution of the complex equations which usually resulted. It has only been in the last 10 to 15 years that mathematical modeling of complex processes has yielded significant accomplishments. Most notable among these have been the simulations used in the aerospace industry, especially in the manned space flight program.

Although employed successfully as a process study tool for a number of years in private industry, the application of chemical process simulation to the U. S. Army Armament Command's explosive and propellant manufacturing processes is just now beginning to be appreciated. For the past three and a half years, the Chemical Process Technology Division of the

Manufacturing Technology Directorate at Picatinny Arsenal has been engaged in an extensive effort to improve, via the application of computer simulation, a continuous process for TNT manufacturing. This work has been carried out jointly with Imperial Chemical Industries, U. S. (ICIUS) the operating contractor at Volunteer Army Munition Plant in Chattanooga, Tennessee, and has to date gone through three distinct phases: Phase I, initial development of a kinetics and mass transfer model of the nitration section with subsequent implementation on a hybrid computer; Phase II, conversion of the hybrid nitration model to a pure digital model, followed by improvement of the nitration model based on new plant data and additional theoretical considerations, simulation of nitration section hydraulics, and development of a simulation of the purification section of the process; and Phase III, extensive exploitation of the nitration and purification computer models.

Project summary reports (Ref 1, 2, 3) have previously been prepared covering these efforts. However, no unified text has been generated which ties together all the interrelated tasks and provides a much needed explanation of the present structure, capabilities, and limitations of various models developed. The purpose of this report is to satisfy these needs as well as to clarify material in the existing reports which has either become outdated or which the author feels is unclear as presently stated.

Because of the extensive amount of information which is presented, the report has been prepared in three volumes. Volume I describes the modeling and simulation effort that has been carried out on the nitration section of the process, including a description of the process chemistry and equipment. Volume II deals exclusively with the purification section model, and Volume III involves the simulation of the hydraulic phenomena occurring in a nitration stage.

### THE CHEMISTRY OF THE NITRATION - A BRIEF DESCRIPTION

The production of TNT is essentially carried out via a three-step reaction in which toluene and its mono- and dinitro-substituted derivatives are contacted with mixtures of nitric and sulfuric ucid called mixed acid. Each step in the reaction corresponds to the successive electrophilic substitution of a hydrogen ion (H<sup>+</sup>) by a nitronium ion (NO<sub>2</sub><sup>+</sup>) on the aromatic ring of the toluene molecule. The nitric acid serves as the source of NO<sub>2</sub><sup>+</sup> ions, while the sulfuric acid acts as both a catalyst and dehydrating agent, first reacting with the nitric acid to generate NO<sub>2</sub><sup>+</sup> ions and then picking up the water which forms as a product of the nitration reaction.

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A simplified scheme for the mononitration of toluene in mixed acid can be written as follows:

$$HNO_3 + H_2SO_4 \longrightarrow NO_2^+ + HSO_4^- + H_2O$$
 (1)

$$CH_3 \qquad CH_3 \qquad NO_2 \qquad + NO_3^+ + HSO_4^- \qquad O-MNT \qquad + H_2SO_4 \qquad (2)$$
(Toluene)

Adding reactions 1 and 2 gives the overall reaction:

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Similarly, when toluene is replaced by mononitrotoluene (MNT) or dinitrotoluene (DNT), the stoichiometric reactions are written as

$$\begin{array}{c}
CH_3 \\
NO_3
\end{array}
+ HNO_3 \xrightarrow{H_2 SO_4}$$

$$\begin{array}{c}
CH_3 \\
NO_3
\end{array}
+ H_3 O$$

$$(4)$$

The temperature as well as the composition of mixed acid required to efficiently carry out each of the above reactions is different, reflecting the relative ease with which nitration is accomplished. Thus, Reaction 3 is usually conducted at relatively low temperatures (40° to 50°C) with weak acid solutions, while reactions 4 and 5 require progressively higher temperatures (60° to 100°C) and stronger acid mixtures to be driven to the desired degree of completion.

The strength of mixed acid is usually determined by the amount of water present. In aqueous media Reaction 1 controls the generation of nitronium ions. However, when nitration is conducted in an anhydrous medium, the acid mixture consists of nitric acid and oleum  $(100\% \, H_2 \, SO_4)$  in which  $SO_2$  dissolved) and the controlling nitronium ion generation equilibria are:

$$HNO_3 + 2H_1S_2O_7 \implies NO_2^+ + HS_2O_7^- + 2H_2SO_4$$
 (6)

$$HNO_3 + HS_2O_7 \implies NO_2^+ + 2HSO_4^-$$
 (7)

It is also worth noting that Reactions 3, 4, and 5 are highly exothermic.

Although not shown in Reactions 3 through 5, a significant amount of isomerism occurs, resulting in the production of a variety of nitrated components in addition to the desired  $\alpha$ -TNT. A complete description of the isomerism in toluene nitration with relative amounts of each component formed is given in Figure 1. The relative amounts of isomers formed depend upon nitrating temperature and acid composition. Thus Figure 1 must be viewed as a typical distribution only. In the continuous TNT process at Radford Army Ammunition Plant, only the  $\alpha$ ,  $\beta$ ,  $\gamma$ , and  $\epsilon$  isomers have been positively identified, although by the analytical technique used on process samples, it is not possible to separate the  $\alpha$  and  $\eta$  isomers (Ref 4).

In addition to its function as a nitrating agent, nitric acid also plays the role of an oxidant under the usual conditions found in toluene nitration reactions. All nitrated components are capable of undergoing some form of oxidation. Two types of oxidation reactions are observed: those which result in destruction of the aromatic ring, yielding gaseous products, and those which result in oxidation of the methyl group or hydroxylation of the aromatic ring, yielding stable organic compounds. Typically, MNT undergoes hydroxylation to the cresol which is later destructively oxidized to gaseous products; DNT undergoes complete ring destruction

to yield CO<sub>2</sub>, CO, NO, NO<sub>2</sub> and tetranitromethane (TNM); and TNT undergoes methyl group oxidation to yield a variety of organic products as shown in Figure 2.

In a typical nitration of toluene to TNT, isomerism and oxidation account for a yield loss of about 8 to 10% based on molar feed of foluene.

### PROCESS DESCRIPTION

The continuous TNT process, which is the subject of the simulation study, is an adaptation of technology developed by Canadian Industries Limited and is currently the only continuous process for manufacture of TNT operating in the United States.

Nitration is carried out in a series of six reaction stages, each consisting of a 425-gallon nitrator and a 150-gallon separator, except for Stages 1 and 3 which have two nitrators. Because of the ratios of mixed acid to toluene employed in the process, a two-phase system results, with nitration occurring in the acid phase as diffusion and solubility phenomena establish equilibrium concentrations of the organic species in that phase. Separation of organic and acid phases is carried out after each nitration stage and enables countercurrent flow of the reactants to be maintained. A flow diagram of the nitration section of the process is shown in Figure 3, and a detailed diagram of a nitrator/separator nitration stage is shown in Figure 4.

In the countercurrent process, all of the toluene is introduced into the first stage, where mononitration and a small amount of dinitration occurs. After separation, this so-called "nitrobody" stream moves forward through the reactor train, becoming more highly nitrated as it progresses from vessel to vessel. Oleum is fed to Stage 6 and by means of the mixed acid recycle streams, moves in the opposite direction, decreasing in strength as it flows toward Stage 1. Nitric acid is added to each stage in order to maintain the correct mixed acid composition. Mixed acid recycle streams also contain some dissolved nitrobody as well as nitrosylsulfuric acid (HNOSO<sub>4</sub>) which forms as a result of the oxidation side reactions.

As pointed out previously, nitration becomes progressively more difficult as the number of nitro groups on the aromatic ring of the substrate increases. Thus, the temperature as well as the mixed acid strength

must increase from Stage 1 to Stage 6. Nominal temperatures for the eight nitrators are 55, 50, 70, 80, 85, 90, 95, and 100°C respectively.

From Figure 4 it is seen that a nitrator is a cylindrically shaped vessel with an agitator fitted inside a draft tube at the center. The agitator provides the required interphase mixing in the nitrator and also creates a head difference between material on either side of the draft tube which in turn provides the motive force for fluid flow. There are no pumps in the system. Rapid internal circulation of material over the nitrator's cooling coils is maintained. There is also a recycle of acid between the separator and its own nitrator (internal recycle). Flow of this stream can be adjusted to maintain proper cooling in the separator or to change the acid-to-nitrobody ratio and residence time in the nitrator.

The acid stream flowing from a separator to the previous nitration stage (external recycle) passes through a weir-like device called a decanter. By adjusting this apparatus, the nitrobody/acid interface in the separator can be regulated to insure adequate phase separation. However, under normal operating conditions of 55 tons per day TNT production, a significant amount of entrainment still occurs, especially in Separator 3.

The crude TNT leaving Separator C flows to the purification section where residual nitrating acid and asymmetrical isomers are removed prior to final packaging of the product.

## DEVELOPMENT OF THE MATHEMATICAL MODEL

## The Single Vessel Model

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The majority of the work conducted under the TNT process computer simulation effort has been devoted to the construction of a mathematical model which describes the nitration section of the process in terms of the kinetic and mass transfer phenomena which are believed to occur there and account for most of the process' behavior. As previously indicated, models of the purification section and of the hydraulic phenomena which govern fluid flow in a given nitration stage have also been developed but will not be discussed in this volume.

The kinetic and mass transfer model of the nitration section of the process (hereafter called the nitration section model) is the end product of numerous refinements and improvements which have been made to a first-generation model developed for Picatinny Arsenal in May 1970 by Kinotrol, Inc. of Houston, Texas, functioning as a consultant to ICIUS at Volunteer Army Ammunition Plant in Chattanooga, Tennessee. The structure and contents of the original nitration section model can be found in Reference 1.

As in the original, a fundamental approach was taken in developing the present nitration section model. Wherever possible, theoretical equations are employed to represent the chemical and physical phenomena which govern the nitration process. The use of empirically derived equations and correlations is kept to a minimum and has been resorted to only after exhaustive research for theoretical alternatives proved fruitless. Phenomena described by the model include kinetics of nitration and oxidation of organic species, diffusion of the organic species between the acid and organic phases, solubility of organic species in the acid phase, and solubility of nitric acid in the organic phase. The way in which real plant data was used in the development of the nitration section model will be described when parameter fitting is explained in a later section of this report.

The easiest way to explain the nitration section model is to take a single vessel in the nitration train (there are 14 vessels in series) and develop a complete material balance for each component in both acid and organic phases. The generalized nitrator in Figure 5 showing actual stream flows could be used for this purpose. However, the analysis is far simpler if each stream is broken down into its acid and organic phases with the subsequent addition of similar phases, so that the vessel input and output consist of a single acid phase, A, a single organic phase P, and a gas phase, G. Figure 6 illustrates this simplifying concept. Thus, for component i in the acid phase, a mole balance can be written as:

(Rate of moles i in with  $A_0$ ) +

(Rate of generation of moles i in the acid phase within nitrator n) -

(Rate of moles i out with  $A_n$ )

(Rate of accumulation of moles i within the acid of nitrator n)

(1)

The balance for component i in the organic phase follows the same form as Equation 1. For the gas phase, the balance is simplified by the fact that products of the gaseous reaction are insoluble in both the acid and organic phases. Thus,

For a specific component, say  $\alpha\text{-TNT}$ , the molar balances for vessel n are written as:

$$A_{n,\alpha-TNT}^{0} + \Sigma S_{n,\alpha-TNT}^{A} - A_{n,\alpha-TNT} = \frac{dA_{n,\alpha-TNT}}{dt}$$
(3)

for the acid phase, where SA is the acid phase generation term, and

$$P_{n,\alpha-TNT}^{o} + \Sigma S_{n,\alpha-TNT}^{P} - P_{n,\alpha-TNT} = \frac{dP_{n,\alpha-TNT}}{dt}$$
 (4)

for the organic phase, where S<sup>P</sup> is an organic phase generation term. The assumption of a perfectly backmixed reactor allows for the equivalency of the reactor contents and the reactor outflow.

Equations similar to 3 and 4 can be written for all the components in each phase for every vessel in the nitration train. In the current version of the nitration section model, 10 chemical components are considered in the organic phase, 14 in the acid phase, and 3 in the gaseous effluent (see Figure 8). Before proceeding further, a few comments on these components are in order. First, all nitrobody components originating from m-MNT have been lumped together in the m-DNT and m-TNT terms in order to simplify the kinetics of the asymmetrical isomer reactions. Secondly, a single term, TNBX, is used to represent all components other than TNB formed from methyl group oxidation of TNT. This includes trinitrobenzyl alcohol, trinitrobenzaldehyde, and trinitrobenzoic acid. Lumping of oxidation products into one term was necessitated by a lack of process data for these components, and is justified considering the small concentration of each component which is present.

Thus, from Equations 3 and 4, it can be seen that for a given vessel the model consists essentially of a series of first order, non-linear, coupled, ordinary differential equations which make up the vessel's material balance.

The way in which the individual vessels are connected and the procedure for generating the flow and composition of the actual process streams from the ideal phases will be discussed at length in later sections of this report.

The heart of the vessel material balance equations and therefore the heart of the nitration section model is the generation terms. These terms are actually equations which represent the kinetic, mass transfer, and solubility processes occurring within a given vessel. For each component balance equation, three generation terms exist: reaction rate  $R_{n,i}$ , diffusion  $(D_{n,i})$ , and bulk mass transport  $(M_{n,i})$ .

Thus Equation 3 can be rewritten as:

$$A_{n,\alpha TNT}^{O} + R_{n,\alpha TNT}^{A} + D_{n,\alpha TNT}^{A} + M_{n,\alpha TNT}^{A} - A_{n,\alpha TNT} = \frac{dA_{n,\alpha TNT}}{dt}$$
(5)

Since it is widely accepted that all reactions occur in the acid phase, it follows that  $R_{n,i}^P = 0$ . Also mass transfer into the acid phase must be at the expense of material in the organic phase so that  $D_{n,\alpha TNT}^P = -D_{n,\alpha TNT}^A$  and  $M_{n,\alpha TNT}^P = -M_{n,\alpha TNT}^A$ . Thus, Equation 4 can be written as:

$$P_{n,\alpha TNT}^{O} - D_{n,\alpha TNT} - M_{n,\alpha TNT} - P_{n,\alpha TNT} = \frac{dP_{n,\alpha TNT}}{dt}$$
(6)

Note that there is no longer a need for acid and organic phase superscripts on the mass transfer terms. Development of the generation terms  $R_{n,i}^A$ ,  $D_{n,i}$ , and  $M_{n,i}$  will now be discussed in detail.

## Reaction Kinetics

Reaction rate expressions have been developed to represent the nitration and oxidation of toluene and its nitro-substituted derivatives at each stage in the reactor train. The stoichiometric reations which are considered in the nitration section model include:

#### Nitration

Toluene + 
$$HNO_3 \longrightarrow aMNT + H_2O$$
 (Reaction 1)

Toluene +  $HNO_3 \longrightarrow mMNT + H_2O$  (Reaction 2)

 $aMNT + HNO_3 \longrightarrow aDNT + H_2O$  (Reaction 3)

 $mMNT + HNO_3 \longrightarrow mDNT + H_2O$  (Reaction 4)

 $aDNT + HNO_3 \longrightarrow aTNT + H_2O$  (Reaction 5)

 $mDNT + HNO_3 \longrightarrow mTNT + H_2O$  (Reaction 6)

#### Oxidation

DNT + 13HNO<sub>3</sub> + 11H<sub>2</sub>SO<sub>4</sub> 
$$\longrightarrow$$
 TNM + 8CO<sub>1.8</sub>  
+ 11HNOSO<sub>4</sub> + 15H<sub>2</sub>O (Reaction 7)  
DNT + 12½HNO<sub>3</sub> + 14½H<sub>2</sub>SO<sub>4</sub>  $\longrightarrow$  7CO<sub>1.8</sub>  
+ 14½HNOSO<sub>4</sub> + 18½H<sub>2</sub>O (Reaction 8)  
HNOSO<sub>4</sub> +  $(\frac{r+2}{3})$  H<sub>2</sub>O + r [O]  $\longrightarrow$  H<sub>2</sub>SO<sub>4</sub>  
+  $(\frac{2-2r}{3})$  NO +  $(\frac{1+2r}{3})$  HNO<sub>3</sub> (Reaction 9)

4TNT + 9HNO<sub>2</sub> + 9H<sub>2</sub>SO<sub>4</sub> 
$$\longrightarrow$$
 TNBOH + TNBAL  
+ TNBA + TNB + CO<sub>2</sub> + 12H<sub>2</sub>O + 9HNOSO<sub>4</sub> (Reaction 10)

(Reaction 9)

It is pointed out that Reactions 1 through 10 are not mechanistic equations but rather stoichiometric equations which account for the appearance and disappearance of the various chemical components during the nitration process.

As mentioned briefly in the section on process chemistry, aromatic nitration in mixed acid occurs via electrophillic substitution, where in most instances the electrophile has been proven to be the nitronium ion.

However, it has also been well established (Ref 8) that as the molar ration of water to sulfuric acid in the mixed acid increases to unity, the

concentration of  $NO_2^+$  falls off rapidly to nearly spectroscopically undetectable levels. Thus, in the case where  $[H_2O]/[H_2SO_4] > 1$  (the weak acid region) and nitration is still seen to occur at a rapid rate, either the aromatic substrate is reactive enough to require only trace amounts of nitronium ion for reaction to proceed, or another nitration mechanism, say one dependent on molecular nitric acid is in effect. The former is more likely but the latter cannot be ruled out.

In the continuous TNT process, all of the monitration and a significant amount of dinitration occurs in mixed acid where  $[H_2O]/[H_2SO_4]>1$ , so that in developing a kinetics model for this weak acid region one of the two mechanism hypothesis must be chosen . It was first decided to test both hypotheses against actual process performance data. While the nitronium ion mechanism was eventually shown to be preferable and was subsequently included in the overall nitration section model, a detailed description at the weak acid model based on molecular nitric acid is nevertheless also presented here for reasons of completeness.

The molecular nitric dependent weak acid kinetics model was based on the work of Kobe and Lakemeyer (Ref 7) and McKinley and White (Ref 8) who investigated the factors affecting the mononitration of toluene in mixed acid where the molar ratio of water to sulfuric acid was greater than two. A mechanism is proposed whereby the nitrosonium ion, NO, is first formed, then attacks the benzene ring and is subsequently oxidized by nitric acid to yield the mononitrated product. This mechanism can be represented as follows:

Although not definitively stated in the literature, Equation 8 appears to be the rate limiting step since data taken by McKinley and White show a strong dependence on the concentration of molecular nitric acid in the acid phase. McKinley and White's nitration studies also indicate that the sulfuric acid concentration has a significant influence on the rate of toluene

nitration in weak acid. Finally, Kobe and Lakemeyer demonstrated that the presence of nitrosylsulfuric acid in weak nitrating acid mixtures causes a maximum in the rate at 4 mole % HNOSO $_4$ .

All of the above observed behavior was combined to yield an expression for the bimolecular rate constant for nitration in weak acid. This expression should be applicable to toluene, MNT, or DNT as long as the conditions of a weak acid nitrating medium are met. The form of this expression is:

$$Rate_{N} = \frac{d[N]}{dt} = k_{2} [HNO_{2}] [M]$$
 (9)

$$k_2 = [H_2 SO_4]^C 10^{-R^2 / (HNOSO_4)^2}$$
 (10)

The dependence of weak acid nitration reactions on temperature was expressed through an Arrhenius-type relationship and, reaction was limited to the acid phase. Thus, Equations 9 and 10 can be restated as:

$$Rate_{N} = \frac{d[N]}{dt} = k_{N} e^{-\frac{E_{n}}{R} \left(\frac{1}{T} - \frac{1}{T_{R}}\right)}$$
 [M] 8 (11)

$$\delta = V_a [HNO_3] [H_3SO_4]^C 10^{-a^2} (X_{HNOSO_4}^{-.04})^2$$
 (12)

where

V is the volume of the acid phase;

 $k_N$  is the Arrhenius frequency factor;

 $\mathbf{E}_{\mathbf{N}}$  is the activation energy; and

 $T_{\mathbf{p}}$  is a reference temperature.

Plant data for the continuous process at Radford indicates that toluene will react to meganitrotoluene just as fast as it can diffuse into the acid phase. Thus for Reactions 1 and 2, mass transfer is the limiting factor and the weak acid kinetics developed above are of academic interest only. However, for conversion of MNT to DNT in weak acid, the weak acid mechanism would apply.

The above model is obviously based on rather unstable theoretically grounds and even though a reasonably good fit to plant data was achieved with it, it was scraped in favor of a more theoretically consistent nitronium ion dependent system of equations.

In this approach the reaction rate is still expressed as in Equation 9; however the expression for  $k_2$  is simply:

$$k_1 = V_n K_N Q \tag{12a}$$

where:

$$K_{N} = k_{N} e^{-\frac{E_{N}}{R}} \left( \frac{1}{T} - \frac{1}{T_{R}} \right)$$
 (12b)

$$Q = \frac{[NO_8^+]}{[HNO_8]} \tag{12c}$$

Utilization of this model depends on the ability to compute the nitronium concentration from the equilibrium reactions which are in effect. The controlling equilibria are chosen to be (Ref 18):

$$HNO_3 + H_2SO_4 \longrightarrow NO_2^+ + HSO_4^- + H_2O$$
 (12d)

$$H_2O + H_2SO_4 \longrightarrow H_2O^+ + HSO_4^-$$
 (12e)

The procedure by which the NOs concentration is calculated follows below.

In the weak acid region, the initial and equilibrium concentrations are given as:

Component	<u>Initial</u>	At Equilibrium	
HNO <sub>3</sub>	[HNO <sub>3</sub> ]	[HNO <sub>a</sub> ]	
H <sub>2</sub> 80 <sub>4</sub>	[H <sub>1</sub> 80 <sub>4</sub> ]	[H <sub>2</sub> SO <sub>4</sub> ]	
H <sub>1</sub> O	(H <sub>1</sub> O)	[H <sub>2</sub> O]	
NO <sub>R</sub>	0	[NO <sub>2</sub> +]	
H₃ O <sup>+</sup>	0	[H <sub>8</sub> O <sup>+</sup> ]	
H80 <sub>4</sub>	0	[HSO <sub>4</sub> ]	

By writing material balances for each chemical element, a system of equations can be developed which, in conjunction with the equilibrium Equations 12d and 12e, can be used to solve for the desired NO<sub>2</sub> concentration at equilibrium. The elemental material balances are based on the necessary equivalence of the moles of chemical elements present initially and the moles of chemical elements present at equilibrium.

For Nitrogen:

$$[HNO3]O = [HNO3] + [NO3+]$$
 (12f)

For Sulfur:

$$[H_1SO_4]_0 = [H_1SO_4] + [HSO_4]$$
 (12g)

For Oxygen:

$$[H_1O]_0 + 3 [HNO_1]_0 + 4 [H_1SO_4]_0 = [H_1O]$$

$$+ 3[I - 4[H_1SO_4] + 2[NO_1^+]$$

$$+ 4[HSO_4] + [H_1O^+]$$
 (12h)

Making use of Equations 12f and 12g in Equation 12h gives:

$$[H_1O]_0 + [NO_1^{\dagger}] = [H_1O] + [H_1O^{\dagger}]$$
 (12i)

For Hydrogen:

$$2[H_{2}O]_{0} + [HNO_{2}]_{0} + 2[H_{2}SO_{4}]_{0} = 2[H_{2}O]$$

$$+ [HNO_{2}] + 2[H_{2}SO_{4}] + [HSO_{4}]$$

$$+ 3[H_{2}O^{+}]$$
 (12j)

Multiplying Equation 12i by 2 and subtracting from Equation 12j gives:

$$[HNO_{3}]_{0} - 2[NO_{3}^{+}] + 2[H_{3}SO_{4}]_{0} = [HNO_{3}]$$
  
+  $2[H_{3}SO_{4}] + [HSO_{4}^{-}] + [H_{3}O^{+}]$  (12k)

And subtracting Equations 12f and g from Equation 12k and gives:

$$\{HSO_{4}^{-}\} = [H_{8}O^{+}] + [NO_{8}^{+}]$$
 (122)

Now, let the initial concentrations be given as:

$$[H_{1}O]_{0} = W$$
 $[H_{1}SO_{4}]_{0} = S$ 
 $[HNO_{1}]_{0} = F$ 

and the equilibrium concentrations of nitronium and bisulfate ions be given as x and y respectively. Then at equilibrium, the concentration of the remaining components can be expressed as:

The equilibrium constant for Equation 12d is given by:

$$K_{1} = \frac{[NO_{2}^{+}] [HSO_{4}^{-}] [H_{2}O]}{[H_{2}SO_{4}] [HNO_{2}]} = \frac{(x) (y) (W+2x-y)}{(s-y) (F-x)}$$
(13)

and that for Equation 12e by:

$$K_{1} = \frac{[H_{1}O^{+}][HSO_{1}]}{[H_{1}O][H_{1}SO_{4}]} = \frac{(y-x)(y)}{(W+2x-y)(S-y)}$$
(14)

Based on the impossibility of negative concentrations, the assumed existence of all possible components at equilibrium, the following constraints must apply:

$$0 < x < y$$
 if  $y < F$   
 $0 < x < F$  if  $F < y$   
 $0 < y < (W+2x)$  if  $(W+2x) < S$   
 $0 < y < S$  if  $S < (W+2x)$ 

Knowing  $K_1$ ,  $K_2$ , F, W, and S, and utilizing the above constraints, Equations 13 and 14 can be solved for x, the nitronium ion concentration at equilibrium. From Equation 14,

$$x = \frac{y^{1} - K_{1} (S-y) (W-y)}{2K_{1} (S-y) + y}$$
 (15)

Equation 15 can be solved for x and y by an interval halving procedure after which the solution values can be substituted in Equation 13 to give

a value of  $K_1$ . If convergence on  $K_1$  is not achieved, the procedure is repeated using the current value of y to adjust the upper or lower boundary of the solution interval for x. In actual execution of the model, between 20 and 40 iterations are required to converge on  $K_1$  for each pass through the vessel equations.

Before moving on to the strong acid region it should be pointed out that a general mechanism for aromatic nitration in aqueous mixed acid has been proposed (Ref 9) based on activity coefficients, but lack of data prevented using it in the nitration section model. An attempt was made, however, to do without any theoretically based weak acid mechanism and just use a curve fit of laboratory data developed by Vinnik (Ref 10) for nitration of p-MNT in mixed acid. The lower end of the curve had to be extrapolated since no data was provided for  $H_2SO_4$  concentrations less than 80%. In all cases, the weak acid mechanism of Equations 9, 12a, 12b and 12c gave far better agreement with observed plant behavior, and it is these equations that constitute the current model.

For strong acid nitration (i.e.  $[H_2O]/[H_2SO_4] < 1$ ) it is firmly established that the nitronium ion is the active nitrating species. Bennett (Ref 11) developed an expression for the bimolecular nitration rate constant,  $k_2$ , in the reaction

$$\frac{d[TNT]}{dt} = k_2 [HNO_2][DNT]$$
 (18)

where the dependence on the  $NO_2^+$  ion concentration as well as the concentration of proton acceptor components ( $HSO_4$ ,  $H_2SO_4$ , and  $HS_2O_7$ ) are considered. Bennett's expression is given as follows:

$$k_2 = Q \left( k' [HSO_4] + k'' [H_2SO_4] k''' [HS_2O_7] \right)$$
 (17)  
where  $Q = \frac{[NO_2^+]}{[HNO_2]}$ 

In order to use Bennett's equation to represent strong acid nitration it is necessary to determine the nitronium and proton acceptor ion concentrations throughout the nitration section of the process. In the model, four strong acid regions are defined in which these ions should exist, and

then equilibrium equations are established which allow calculation of the ionic component concentrations. The first two regions are exactly those dealt with by Bennett. However the third and fourth regions have apparently never been examined experimentally so that the hypothesis presented below constitutes an extension of existing theory.

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The first region (aqueous region) is characterized by the presence of water subject to the constraint  $[H_1O]/[H_2SO_4] < 1$ . The equilibria established in this situation are exactly those of Equations 12d and 12e for which the procedure to determine the equilibrium concentrations has been given.

The remaining three regions are water free, with nitric, sulfuric, and pyrosulfuric  $(H_1S_1O_7)$  acids making up the nitrating acid. These will be referred to as oleum regions.

In the first cleum region (nitric limited), pyrosulfuric acid is in large excess of the nitric acid to the extent that

$$[HNO_3] \leq .5 [H_1S_2O_7]$$

This is the strong oleum region described by Bennett. The equilibria governing concentrations in this region are:

$$HNO_{2} + 2H_{2}S_{2}O_{7} \longrightarrow NO_{2}^{+} + HS_{2}O_{7}^{-} + 2H_{2}SO_{4}$$
 (18)

$$H_1SO_4 + H_2S_2O_7 \longrightarrow H_3SO_4^+ + HS_2O_7^-$$
 (19)

Equation 18 is believed to go completely to the right, thus leaving Equation 19 as the limiting equilibrium. A two-step reaction occurs, the nitric acid first completely dissociating to nitronium ions via Equation 18, followed by the establishment of equilibrium among sulfuric, pyrosulfuric acids and their ionic counterparts via Equation 19. The concentrations are represented as follows:

	Initial	After 18	After 19 (Equilibrium)
HNO <sub>3</sub>	[HNO <sub>3</sub> ]	0	0
H2S2O7	[H2S2O7]	$[H_2S_2O_7]_0$ -2 $[HNO_3]_0$	$[H_1S_2O_7]_0 - 2[HNO_3]_0 - Y$
H <sub>2</sub> SO <sub>4</sub>	[H <sub>2</sub> SO <sub>4</sub> ]	[H <sub>2</sub> SO <sub>4</sub> ] +2[HNO <sub>3</sub> ]	$[H_{1}SO_{4}]_{0} + 2[HNO_{1}]_{0} - Y$
HS, 0,	0	[HNO,]	[HNO <sub>3</sub> ] + Y
NO3	0	(HNO,)	[HNO;]
H.SO4	0	0	Y

where Y is the concentration of  $HS_2O_7^-$  formed via Equation 19. The mass action expression for Equation 19 is:

$$K_{eq} = \frac{[H_1SO_4^+] [HS_1O_7^-]}{[H_1SO_4] [H_1S_1O_7]}$$
(20)

Substitution of the tabulated values yields:

$$K_{eq} = \frac{Y([HNO_3]_o + Y)}{([H_3SO_4]_o + 2[HNO_3]_o - Y)([H_3S_3O_7]_o - 2[HMO_3]_o - Y)}$$
(21)

If  $K_{eq}$  and initial concentrations are known, Y can be found by successive iterations of Equation 21. Bennett has suggested a value of 1.0 for  $K_{eq}$ .

In the second oleum region (intermediate), the pyrosulfuric acid is in moderate excess of the nitric such that:

$$.5[H_2S_2O_7] < [H_1,] \le [H_2S_2O_7]$$

The dominant equilibria in effect here are (Ref 19):

$$HNO_3 + 2H_2SO_7 \longrightarrow NO_3^+ + HS_2O_7^- + 2H_2SO_4$$
 (22)

$$HNO_3 + 2H_2SO_4 \longrightarrow NO_3^+ + H_3O^+ + 2HSO_4^-$$
 (23)

$$H_{3}O^{+} + HS_{3}O_{7}^{-} \longrightarrow 2H_{3}SO_{4}$$
 (24)

Again it is assumed that Equation 22 goes completely to the right. Because of the limits imposed on the nitric acid concentration, it must be true that the nitric acid remaining undissociated after reacting with  $H_1S_2O_7$  (Eq 22) must always be less than the  $HS_1O_7$  formed by the same reaction. Thus, there will always be enough  $HS_2O_7$  present to soak up all of the  $H_2O_7$  formed via equilibrium Equation 23. This continuously drives Equation 23 to the right until all the remaining nitric acid is completely converted to nitronium ions. Equations 23 and 24 can be added to give

$$HNO_{2} + HS_{2}O_{7}^{-} \longrightarrow NO_{2}^{+} + 2HSO_{4}^{-}$$
 (25)

In effect a two-step reaction occurs. Nitric acid reacts with the pyrosulfuric acid via Equation 22, and then the remaining undissociated nitric acid reacts completely with the  $HS_2O_7$  produced from the first reaction. Both reactions go to completion and no equilibria are in effect. Concentrations are as follows:

	Initial	After 22	After 25
H <sub>2</sub> S <sub>2</sub> O <sub>7</sub>	[H <sub>2</sub> S <sub>2</sub> O <sub>7</sub> ]	0	0
H <sub>2</sub> SO <sub>4</sub>	[H2SO4]	[H2SO4] + [H2S2O7]	$[H_1SO_4]_0 + [H_1S_1O_7]_0$
NO <sub>3</sub>	0	.5[H <sub>2</sub> S <sub>2</sub> O <sub>7</sub> ]	[HNO,]
HS <sub>2</sub> O <sub>7</sub>	0	.5[H <sub>2</sub> S <sub>2</sub> O <sub>7</sub> ]	$[H_{1}S_{1}O_{7}]_{0} - [HNO_{1}]_{0}$
HSO.	0	0	2[HNO <sub>3</sub> ] - [H <sub>2</sub> S <sub>2</sub> O <sub>7</sub> ]
HNO;	[HNO,]	$[HNO_{1}]_{0}5[H_{1}S_{1}O_{7}]_{0}$	0

While Gillespie maintains that Equation 25 is completely to the right, there is some basis for the occurrance of the reverse reaction (Ref 20). For this reason, the model has been structured so that, as an option, Equation 25 can be treated as an equilibrium with

$$K_{eq} = \frac{[HNO_{2}][HS_{2}O_{7}^{-}]}{[NO_{2}^{+}][HSO_{7}^{-}]^{2}}$$
(25a)

Initial concentrations are assumed to be those existing after Equation 25 has gone completely to right at which time  $NO_2$  and  $HSO_4$  begin to recombine via the reverse reaction. Equilibrium concentrations are found by assuming a value for  $K_{eq}$  and solving Equation 25a by interval halving.

In the third oleum region (oleum limited), nitric acid exists in excess of pyrosulfuric acid, i.e.  $[HNO_2] > [H_2S_2O_7]$ . The same equilibria are in effect here as in the intermediate region, but now Equation 25 can apply only until the  $HS_2O_7$  produced by Equation 22 is completely dissipated. There will still be some undissociated nitric remaining which must then undergo equilibrium dissociation via Equation 23. In effect, a three-step reaction takes place: 1) Nitric acid  $\rightarrow NO_2$  in the presence of  $H_2S_2O_7$ . 2) Nitric acid  $\rightarrow NO_2$  via Equation 25 until all the  $HS_2O_7$  is dissipated, and 3) Equilibrium dissociation of the remaining nitric acid via Equation 23. Concentrations are as follows:

	Initial	After 22	After 25	After 23 (Equilibrium)
HNO <sub>3</sub>	[HNO,]	[HNO; ]5[H; S; O; ]	[HNO <sub>1</sub> ] <sub>0</sub> - [H <sub>1</sub> 8 <sub>1</sub> O <sub>1</sub> ] <sub>0</sub>	[HNO <sub>2</sub> ] - [H <sub>2</sub> S <sub>2</sub> O <sub>7</sub> ] - 1
H, S, O,	[H2S2O7]	0	0	0
H <sub>1</sub> 80 <sub>4</sub>	[H <sub>2</sub> BO <sub>4</sub> ]	[H2SO4] + [H2S2O7]	[H:SO4] + [H:S:O,]	[H <sub>1</sub> SO <sub>4</sub> ] + [H <sub>1</sub> S <sub>1</sub> O <sub>7</sub> ] - 2x
NO.	0	.5[H <sub>1</sub> S <sub>1</sub> O <sub>7</sub> ]	(H,S,O,)	(H <sub>8</sub> 8 <sub>2</sub> O <sub>7</sub> ) <sub>0</sub> + 2
HS <sub>1</sub> O <sub>7</sub>	0	.5[H <sub>2</sub> S <sub>2</sub> O <sub>7</sub> ]	0	0
HSO.	0	0	[H <sub>1</sub> S <sub>1</sub> O <sub>7</sub> ]	$[H_{1}S_{2}O_{7}]_{O} + 2z$
H₃O <sup>+</sup>	0	0	0	2

where z is the concentration of nitronium ions formed via the equilibrium Equation 23.

As before, the mass action expression for Equation 23 is:

$$K_{eq} = \frac{(NO_{2}^{+})[H_{2}O_{4}^{+}]^{2}}{[HNO_{2}][H_{2}SO_{4}]^{2}}$$
(28)

Substitution of the appropriate tabulated values into Equation 26 gives:

$$K_{eq} = \frac{([H_1S_1O_7]_0 - z)(z)([H_1S_2O_7]_0 + 2z)^2}{([HNO_3]_0 - [H_2S_2O_7]_0 - z)([H_2SO_4]_0 + [H_2S_2O_7]_0 - 2z)^2}$$
(27)

If  $K_{eq}$  and the initial concentrations are known, z can be found by successive iteration of Equation 27.

As was done for the second oleum region, provision has been made to consider recombination of  $NO_2$  and  $HSO_4$  via Equation 25. If this option is chosen in the model, Equations 23 and 25a must be solved simultaneously to give the equilibrium concentrations of the various ionic species. A procedure analogous to that used in the weak acid region to solve Equations 12d and 12e is employed.

Now, knowing initial concentrations, and having solved for the appropriate concentrations at equilibrium, Bennett's proposed rate expression can be applied to strong acid nitration as follows:

$$\frac{d[N]}{dt} = (k'[HSO_4] + k''[H_3SO_4] + k'''[HS_3O_7]) [M][HNO_3]_0 Q$$
(28)

where Q is the fraction of nitric acid existing as nitronium ions;

[M] is the concentration of nitrotoluene reactant:

[N] is the concentration of nitrotoluene product; and k , k , and k are rate constants.

It is emphasized that Equation 28 is continuous over the four strong acid regions previously described.

The influence of temperature upon reaction rate is taken into account by assuming, as Bennett did, that the constants k, k, and k exhibit an Arrhenius dependence.

Thus, Equation 28 can be rewritten as:

$$\frac{d[N]}{dt} = k_{N} e^{-\frac{E_{N}}{R}} \left(\frac{1}{T} - \frac{1}{T_{R}}\right) (k_{B} [HSO_{4}] + k_{D} [H_{2}SO_{4}] + k_{C} [HS_{2}O_{7}]) [M] [HNO_{2}]_{Q} Q$$
(29)

and Equation 29 can be further simplified,

Rate<sub>N</sub> = k<sub>N</sub> e 
$$\frac{-E_N}{R} \left( \frac{1}{T} - \frac{1}{T_R} \right)$$
 [M]  $\gamma$  (30)

$$\gamma = V_a (k_a [HSO_4] + k_b [H_BSO_4] + k_c [HS_BO_7]) [HNO_3]_Q (31)$$

where  $k_N$  and  $E_N$  are the velocity coefficient and activation energy associated with the nitration reaction:

$$M + HNO_3 + N + H_1O$$

and  $V_a$  is the volume of the acid phase.

Subsequent to Bennett's work, strong experimental evidence was uncovered which contradicted a portion of his basic hypothesis. Melander demonstrated that proton departure from the transition compound during nitration is kinetically insignificant (Ref 12). Therefore, Bennett's contention that DNT nitration in mixed acid is a termolecular process dependent on both the nitronium ion and the proton acceptor ions, is most likely incorrect. In addition, for two-phase nitration systems, no maximum in

the rate at 92% HaSO4 has been noted.

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Returning to Equations 18 and 17, it appears that there is sufficient justification to eliminate the proton acceptor component concentration terms from the expression for  $\mathbf{k_1}$  so that

$$k_{2} = k \frac{[NO_{2}^{+}]}{[11.5O_{2}]}$$
 and (31a)

$$\gamma = V_{g} k_{2} [HNO_{3}]$$
 (31b)

with the dependence of nitration on the concentrations of  $SO_3$ ,  $H_2SO_4$ , and  $H_2O$  considered only in so far as those concentrations affect nitronium ion equilibria.

Although the nitration section simulation has been modified to utilize this simpler strong acid nitration model, the option to use Bennett's mechanism has been retained.

Far less is known about the kinetic mechanisms that govern the oxidation reactions which always occur to some extent during TNT manufacture along with the desired nitration reactions. With regard to oxidation, three key assumptions are made in the model: (1) oxidation reactions occur exclusively in the acid phase and therefore compete with the nitration reactions, (2) the controlling component in oxidation is the same as that which governs nitration (NO<sub>2</sub> ion), and (3) oxidation reactions exhibit bimolecular kinetics and are thus subject to the same reaction mechanisms as nitration. Also, because of a lack of plant data on cresol formation, oxidation of MNT is not represented in the model. This however, is not a critical shortcor, a since cresols are eventually destructively oxidized to gaseous products and their contribution to overall yield loss can be compensated for by adjustment of the DNT oxidation rates. Equations A-42 through A-55 are employed in the model to represent oxidation.

## Diffusion

Diffusion, as employed in the nitration section model, is the process by which the nitrobody components move by tween the two phases after their acid phase saturation solubility has been reached. This phenomenon depends solely upon the concentration gradients of the nitrobody species across each phase and on the fluid properties of the medium as reflected by the mass transfer coefficient.

At steady state, diffusion of components toward the acid/organic interface must be equal to that away from it.

Thus,

$$D_{i} = \eta_{0} (y_{i} - y_{i}^{*}) = \eta_{A} (x_{i}^{*} x_{i})$$
 (32)

where

$$x_i = \frac{A_i}{A_T}$$
 and  $y_i = \frac{\varphi_i}{\varphi_T} \approx \frac{\varphi_i}{\varphi_{nb}}$  (33)

(See Figure 8 for an explanation of the nomenclature.)

Now, if we assume that the acid phase is always saturated with nitrobody,

$$A_{nb} = X_{eq}^{A} A_{T}$$
 (34)

$$A_{T} = \frac{A_{NS}}{X_{eq}^{A}}$$
 (35)

so that by substitution of Equation 35 into Equation 33 we get

$$x_i = \frac{A_i}{A_{nh}} \quad X_{eq}^{A} \tag{36}$$

At the interface, the two phases must be in equilibrium. The equilibrium relationship assumed to be in effect is that the mole fraction nitrobody component in the organic phase must be equal to the acid-free mole fraction of that component in the acid phase.

Thus, at the interface,

$$\frac{\varphi_i^*}{\varphi_{nb}^*} = \frac{A_i^*}{A_{nb}^*} \tag{37}$$

but  $A_{nb}^* = X_{eq}^A A_T^*$ 

so that  $\frac{\varphi_i^*}{\varphi_{nb}^*} = \frac{A_i^*}{X_{eq}^A A_T^*}$  (38)

or 
$$y_i^* = x_i^*/X_{eq}^A$$
 (39)

Substitution of the expressions for  $y_i$  and  $y_i^*$  into Equation 32 gives

$$D_{i} = \eta_{\varphi} \left( \frac{\varphi_{i}}{\varphi_{nb}} - \frac{x_{i}^{*}}{X_{eq}^{A}} \right) \tag{40}$$

but also from Equation 32,

$$x_i^* = \frac{D_i}{\eta_A} + x_i \tag{41}$$

so that

\*

$$D_{i} = \eta_{\varphi} \left[ \frac{\varphi_{i}}{\varphi_{nh}} - \frac{\left(\frac{D_{j}}{\eta_{A}} + x_{i}\right)}{X_{eq}^{A}} \right]$$
 (42)

$$D_{i} = \eta_{\varphi} \left( \frac{\varphi_{i}}{\varphi_{nb}} - \frac{D_{i}}{\eta_{A} X_{eq}^{A}} - \frac{x_{i}}{X_{eq}^{A}} \right)$$
 (43)

$$\frac{D_i}{\eta_{\varphi}} + \frac{D_i}{\eta_{A} X_{eq}^{A}} = \left(\frac{\varphi_i}{\varphi_{nb}} - \frac{X_i}{X_{eq}^{A}}\right) = \left(\frac{\varphi_i}{\varphi_{nb}} - \frac{A_i}{A_T X_{eq}^{A}}\right)$$

(44)

$$D_{i}\left(\frac{1}{\eta_{\phi}} + \frac{1}{\eta_{A}X_{eq}}A\right) = \frac{\varphi_{i}}{\varphi_{nb}} - \frac{A_{i}}{A_{nb}}$$
 (45)

Now let 
$$\frac{1}{\eta} = \left(\frac{1}{\eta_{\varphi}} - \frac{1}{\eta_{A} X_{eq}^{A}}\right) \tag{46}$$

so that 
$$D_{i} = \eta \left( \frac{\varphi_{i}}{\varphi_{nb}} - \frac{A_{i}}{A_{nb}} \right) \tag{47}$$

Equation 47 is true for all organic components except toluene. For toluene, the reaction to MNT is so fast that it is assumed to occur at the interface, which eliminates the acid phase diffusion of toluene and forces  $A_{Tol}$  to be zero. Also the extremely rapid movement of toluene to the interface warrants a separate mass transfer coefficient. Thus,

$$n_{Tol} = \eta_{Tol} \frac{\varphi_{Tol}}{\varphi_{nb}}$$
 (48)

As indicated previously, the rate of conversion of toluene to MNT via Reactions 1 and 2 is solely dependent upon the rate of toluene diffusion so that

$$R_1 + R_2 = D_{Tol}$$

$$\frac{R_1}{R_2} = \frac{D_{Tol}}{R_2} - 1 \tag{49}$$

However, the ratio of aMNT to mMNT produced is dependent on the kinetic expressions:

$$\frac{\alpha MNT}{mMNT} = \frac{R_1}{R_8} = \frac{k_1 \cdot e^{-\frac{R_1}{R}} \left(\frac{1}{T} - \frac{1}{T_R}\right)}{k_8 \cdot e^{-\frac{R_8}{R}} \left(\frac{1}{T} - \frac{1}{T_R}\right)}$$
(50)

$$\frac{R_1}{R_8} = \left\langle \frac{k_1}{k_2} \right\rangle_{\theta} + \left[ -\frac{E_1}{R} \left( \frac{1}{T} - \frac{1}{TR} \right) + \frac{E_8}{R} \left( \frac{1}{T} - \frac{1}{T_R} \right) \right]$$
(51)

$$\frac{R_1}{R_2} = \frac{k_1}{k_2} = \frac{(E_1 - E_2)}{R} = \frac{1}{T} = \frac{1}{T_R}$$
 (52)

$$\frac{R_1}{R_2} = k_{12} e^{-\frac{(E_1 - E_2)}{R}} (\frac{1}{T} - \frac{1}{T_R})$$
 (53)

Substitution of Equation 49 into Equation 53 gives

$$\frac{D_{Tol}}{R_{1}} - 1 = k_{1}, \quad e^{-\left(\frac{E_{1} - E_{2}}{R}\right)} \left(\frac{1}{T} - \frac{1}{T_{R}}\right) \tag{54}$$

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$$\frac{\frac{1}{R_1} = \frac{\frac{1 + k_{12} e}{D_{Tol}} \left(\frac{1}{T} - \frac{1}{T_R}\right)}{\frac{1}{R_1}}$$
 (55)

Now. since

$$k_{13} = 0 - \frac{(E_1 - E_3)}{R} = (\frac{1}{T} - \frac{1}{T_R}) >>> 1$$
, we can re-

write Equation 55 as

$$R_{2} = \frac{D_{Tol}}{k_{12} - \frac{(E_{1} - E_{2})}{R} \left(\frac{1}{T} - \frac{1}{T_{R}}\right)}$$
(56)

$$R_{1} = \frac{D_{Tol}}{k_{10}} e^{-(E_{1} - E_{3})/R(\frac{1}{T} - \frac{1}{T_{R}})} = \frac{D_{Tol}}{k_{10}} e^{-\frac{(E_{3} - E_{1})/R(\frac{1}{T} - \frac{1}{T_{R}})}{(57)}}$$

At this point, it should also be noted that while chemical reactions occur in both the nitrators and separators due to the ever present solubilised organic components in the acid phase, diffusion is assumed to be nonexistent in the separators because of the drastic reduction in interface area which occurs there.

# **Bulk Mass Transfer and Solubility**

The remaining generation terms in the material balance equations (Equations 5 and 6) are those for bulk mass transfer. Bulk mass transfer, is in effect, the means by which equilibrium nitrobody solubility is enforced on the acid phase. At any point in the nitration section, the equilibrium amount of nitrobody in the acid phase is given by

$$A_{nb} = X_{eq}^{A} A_{T}$$
 (58)

where  $X_{eq}^{A}$  has a value of pendent on the local temperature as well as the local acid phase composition. For a given vessel, nitrobody is present in the acid phase by virtue of that which entered with the incoming acid phase,  $A_{nb}^{O}$ , that which diffused in from the organic phase, i=nb  $D_{i}$ , and that which resulted from a change in solubility created by combination of various vessel input streams which are obviously not in equilibrium. The last of these is considered the bulk mass transfer. Thus, to enforce equilibrium on the acid phase leaving the vessel, it must be true that

$$X_{eq} \stackrel{A}{=} A_{T} = A_{nb}^{o} + \sum_{i = nb} D_{i} + M_{nb}$$
 (59)

and

$$M_{nb} = X_{eq}^{A} A_{T} - A_{nb}^{O} - \sum_{i=nb} D_{i}$$
 (60)

where  $M_{nb}$  represents the bulk movement of nitrobody into the acid phase which must occur in order to maintain equilibrium solubility. Negative values of  $M_{nb}$  are reasonable and indicate nitrobody dropping out of solution at the new vessel conditions. The individual component bulk mass transfer rates are computed from the overall bulk nitrobody transfer rate as follows:

$$M_{i} = \begin{cases} \frac{A_{i}}{A_{nb}} & M_{nb} & \text{if } M_{nb} < 0 \\ \\ \frac{\varphi_{i}}{\varphi_{nb}} & M_{nb} & \text{if } M_{nb} \ge 0 \end{cases}$$

where the index i represents each nitrobody component. The change in acid phase nitrobody composition due to reaction is not included in the bulk mass transfer term since reaction only converts one nitrobody component to another and does not result in a net molar change.

An expression for the equilibrium solubility of bulk nitrobody in the acid phase has been developed by subjecting a limited quantity of published solubility data to a multiple regression procedure.\* Solubility is correlated against temperature and acid phase composition to give:

$$x_{eq}^{A} = 10^{p} \tag{61}$$

where

$$p = 3.62 + .01023T - 3.808 \times_{H_8O} + 7.716 \times_{MNT}$$

$$-8.452 \times_{TNT} - 6.292 \times_{HNO_3}$$
(62)

<sup>\*</sup>Laboratory solubility data for TNT, DNT, and MNT in mixed acid at various temperatures is currently being generated. This data, being more extensive than what is used for the existing solubility correlation, will be regressed to generate a new nitrobody solubility expression which is expected to significantly improve the nitration section model.

where

T is temperature in oK

 $x_{H_{\bullet}O}$  is mole fraction water in acid phase

 $\mathbf{x}_{\mathbf{MNT}}$  is mole fraction MNT in acid phase

 $\mathbf{x}_{\mathbf{TNT}}$  is mole fraction TNT in acid phase

\*HNO, is mole fraction nitric acid in acid phase

Finally, an expression has been developed to represent the solubility of nitric acid in the organic phase. It is the only acid component that shows any appreciable solubility in the organic phase. The expression used in the model is:

$$y_{HNO_8} = 1.51 X_{H_8O} \left( \frac{x_{HNO_8}}{x_{H_8SO_4}} \right)$$
 (63)

Expressions for all of the terms in the vessel material balance equations (Equations 5 and 6) have now been developed so that if all vessel inputs are known and can be divided into acid and organic phase components, simultaneous solution of the vessel equations will give the output of that vessel in terms of moles per hour each component in each phase.

The vessel equations can be solved either for the transient state, in which case integration of the first order ordinary differential equations is required, or for the steady state, in which case the derivative terms are set equal to zero, resulting in a system of non-linear, simultaneous, coupled, algebraic equations. Both of these cases have been solved and the solution procedures including the computer programs generated will be described in detail in subsequent sections of this report.

# Formulation of the Overall Nitration Section Model from the Single Vassel Model

Up to this point, discussion has centered around the details of the single vessel model without any regard to the way in which the complete 14-vessel nitration train (8 nitrators and 6 separators) is actually simulated. "Connection" of the individual vessels in the nitration section is

made possible by use of a physical separation model which takes the ideally separated organic and acid phase computed by the separator vessel model and constructs the three real streams which actually flow to other vessels. These include the organic stream forward to the next nitrator, the internal acid recycle stream, and the external acid recycle stream to the previous nitration stage. Because the separation process is not ideal, a portion of the acid phase is entrained in the organic stream leaving the separator and similarly, a portion of the organic phase is entrained in the two exiting acid streams.

In developing the equations for the physical separation model, the entrainments are defined as follows:

The state of the s

- e oa = molar fraction of the acid recycle stream, both internal and external, which is entrained organic phase.
- e molar fraction of the organic stream which is entrained acid phase.

The following assumptions are also made: (1) the composition of entrained material is the same as that of the bulk phase from which it originated, (2) the composition of the internal and external recycle streams are identical, and (3) the values for e and e are constant for a given separator.\*

The separation process is described schematically in Figure 9. It is again stressed that this physical separation model operates on the contents of a separator after the kinetic and mass transfer processes in that separator have transpired.

<sup>\*</sup>An option exists in the model whereby e and e can be computed based on correlations that were developed using linear multiple regression techniques. However, because of the relatively high standard error of estimate associated with the correlation coefficients, e and e are usually assigned their constant values.

Now, let

A, = moles/hr of acid phase in Stream i

 $\phi_i$  = moles/hr of organic phase in Stream i

Note that the flow rates  $A_o$  and  $\phi_o$  of the acid and organic phases in the input stream are known.

First consider the separation of Stream 0 into Streams 1 and 2. From the definition of  $e_{oa}$  and  $e_{ao}$ :

$$e_{\text{oa}} = \frac{\varphi_1}{A_1 + \varphi_1} \tag{64}$$

$$e_{OR} = \frac{A_2}{A_2 + \phi_2} \tag{65}$$

These can be converted into molar ratios as follows:

$$e_{0a}^{'} = \frac{\varphi_1}{A_1} = \frac{e_{0a}}{1-e_{a0}} = \frac{e_{0a}}{1-e_{$$

$$e_{0a}^{'} = \frac{A_2}{\phi_2} = \frac{e_{ao}}{1-e_{ao}} = \frac{e_{ao}}{1-e_{ao}} = \frac{e_{ao}}{e_{ao}} = \frac{e_{ao}}{e_{$$

Material balances around node Ayield:

$$\varphi_0 = \varphi_1 + \varphi_2 \tag{68}$$

$$A_0 = A_1 + A_2 \tag{69}$$

Substituting Equation 66 for  $\phi_1$  and Equation 67 for  $A_2$  gives:

$$\varphi_{O} = A_{1} e_{OB}^{\dagger} + \varphi_{2} \tag{70}$$

$$\mathbf{A}_{\mathbf{O}} = \mathbf{A}_{1} + \mathbf{\varphi}_{2} \mathbf{e}^{\dagger}_{\mathbf{OB}} \tag{71}$$

Eliminating  $\phi_1$  and rearranging, gives

$$f_{ar} = \frac{A_1}{A_2} = \frac{1-e^{i}_{ao} \varphi_{o}/A_{o}}{1-e^{i}_{ao} e^{i}_{oa}} =$$
fraction of the entering acid phase that appears in the acid recycle stream. (72)

Solving Equation 70 for  $\phi_{2}$  , substituting Equation 72, and rearranging gives:

$$f_{po} = \frac{\varphi_a}{\varphi_o} = 1 - f_{ar} e'_{oa} A_o/\varphi_o$$
 (73)

From  $f_{\mbox{\footnotesize po}}$  and  $f_{\mbox{\footnotesize ar}}$  the composition of each component in the organic stream can readily be calculated. Let  $A_{ij}$  be the moles/hr of Component j in the acid phase of Stream i. Define  $\phi_{ij}$  analogously to be the moles/hr of Component j in the organic phase of Stream i. For Stream 2, these can be computed as follows:

$$A_{ij} = (1-f_{ar}) A_{oj}$$
 (74)

$$\varphi_{ij} = f_{po} \varphi_{oj} \tag{75}$$

Stream 1 must now be separated into its internal and external recycle portions. The volumes of the entering phases (Stream 0)

can be computed from their compositions:

$$Q_{\phi 0} = ft^3/hr$$
 of organic phase in Stream 0  
 $Q_{A0} = ft^3/hr$  of acid phase in Stream 0  
 $Q_{T0} = Q_{\phi 0} + Q_{A0} = total ft^3/hr$  for Stream 0.

The total flow of Stream 1 can be computed as:

$$Q_{T_1} = f_{ar}Q_{Ao} + (1-f_{po})Q_{\phi o}$$
 (78)

The fraction of Stream 1 that appears in Stream 3 (internal acid recycle) is:

$$f_{ir} = \frac{Q_{Ta}}{Q_{Ti}} = \frac{Q_{max} f(N)}{f_{ar} Q_{Ao} + (1-f_{po}) Q_{\varphi o}}$$
 (77)

where  $Q_{max} = maximum ft^3/hr$  of internal recycle (a known constant),

N = notch setting on a gate valve in the internal recycle line which is modeled as an equal percentage valve,

f(N) = fraction of maximum flow corresponding to the notch
setting.

The flow rates of the individual components in each phase of Streams 3 and 4 can be computed as follows:

$$A_{ij} = f_{ir} f_{ar} A_{oj}$$
 (78)

$$\varphi_{ij} = f_{ir} (1-f_{po}) \varphi_{oj}$$
 (79)

$$A_{4j} = (1-f_{ir}) f_{ar} A_{oj}$$
 (80)

$$\phi_{4j} = (1-f_{ir}) (1-f_{po}) \phi_{oj}$$
(81)

Thus it is seen that the composition of the separator outlet streams can be calculated by the physical separation model from the results of the separator vessel model and that this in turn forms the basis for connection of the six nitration stages. The way in which the stages are connected will become clearer in the next section of the report, which deals with the implementation of the vessel and physical separation models on a digital computer to form the complete nitration section simulation.

# COMPUTER IMPLEMENTATION OF THE MATHEMATICAL MODEL THE SIMULATION

Separate computer programs have been prepared to simulate both the dynamic and steady states of the nitration process. While differing in solution procedure, there is no difference in their basic structures; both are ways to solve the vessel equations to give vessel outputs for known inputs. In fact, the solution obtained by running the dynamic simulation long enough to reach steady state is exactly that obtained by executing the steady state simulation, within the limits of numerical accuracy. A complete list of the vessel equations (the mathematical model of any nitration section vessel) ordered according to a logical solution sequence is given in Appendix A.

## The Dynamic Simulation

The dynamic simulation is a computer program which, for a given set of nitration section independent variables including (1) feed rates (lb/hr), (2) composition of raw materials (wth), (3) nitrator temperatures (°C), and (4) recycle notch settings, will operate upon the vessel and physical separation equations to give the composition and flow rate (moles/hr) of the exiting streams from each of the 14 process vessels as functions of time. A numerical integration technique known as Euler's Method is employed to solve the differential equations. Although this is the simplest of the many numerical integration algorithms available, its efficiency in solving the vessel equations is satisfactory.

# Formulation of the Equations

In order to solve the unsteady state vessel equations via Euler's Method, it is first necessary to reformulate the continuous differential equations (Equations 5 and 6) into their discrete numerical equivalents.

For Component 1 (aMNT)\*, Equation 5 is

$$A_{n,i}^{0} + R_{n,i}^{A} - R_{n,s}^{A} + D_{n,i} + M_{n,i} - A_{n,i} = \frac{d A_{n,i}}{dt}$$
 (82)

and Equation 6 is

Ū

$$P_{n,1}^{0} - D_{n,1} - M_{n,1} - P_{n,1} = \frac{dP_{n,1}}{dt}$$
 (83)

Equations 82 and 83 are not exactly correct since the units of all terms on the left side are moles/hr while those on the right are moles/ $hr^8$ . To correct for this, the derivatives are rewritten as:

$$\frac{d}{dt}$$
  $(V_n f_A \rho_A x_{n,1})$  for  $\frac{d A_{n,1}}{dt}$ 

and

$$\frac{d}{dt} (V_n (1-f_A) \rho_p y_{n,1}) \text{ for } \frac{d P_{n,1}}{dt}$$

where  $V_n$  is the vessel volume (ft<sup>3</sup>)  $f_A \text{ is the volume fraction acid phase in the vessel}$   $\rho_A \text{ is the density of the acid phase (moles/ft<sup>3</sup>)}$   $\rho_P \text{ is the density of the organic phase (moles/ft<sup>3</sup>)}$   $x_1 \text{ is the mole fraction of } \alpha \text{ MNT in the acid phase}$   $y_1 \text{ is the mole fraction of } \alpha \text{ MNT in the organic phase}.$ 

Therefore Equations 82 and 83 can be rewritten as:

$$A_0 + R_1 - R_3 + D_1 + M_1 - A_1 = \frac{d}{dt} (Vf_A \rho_A x_1) \otimes \frac{V_A f_A \rho_A}{A_T} \frac{d A_1}{dt}$$
 (84)

See Appendix A for component number assignments

$$P_{0} - D_{1} - M_{1} - P_{1} = \frac{d}{dt} \left[ V (1-f_{A}) \rho_{p} y_{1} \right] = \frac{V (1-f_{A}) \rho_{0}}{A_{T}} \frac{dP_{1}}{dt}$$
(85)

Note that in Equations 84 and 85 the vessel subscripts have been dropped and the assumption that fraction acid, density, and total moles change little with time has been made.

The numerical approximations for the continuous derivatives are:

$$\frac{dA_1}{dt} = \frac{A_1^{t+\Delta t} - A_1^t}{\Delta t}$$
 (86a)

$$\frac{d P_i}{dt} = \frac{P_i^{t+\Delta t} - P_i^t}{\Delta t}$$
 (86b)

where the superscript t represents a quantity evaluated at the current time and  $t+\Delta t$  indicated computation at the end of a time step. Substitution into Equations 84 and 85 yields:

$$[A_1^0 + R_1 - R_2 + D_1 + M_1 - A_1]^{\frac{t}{2}} = \frac{Vf_A \rho_A}{A_T \Delta t} [A_1^{t+\Delta t} - A_1^{t}]$$
 (87)

$$[P_1^0 - D_1 - M_1 - P_1]^t = \frac{V(1-f_A) \rho_P}{P_T \Delta t} [P_1^{t+\Delta t} - P_1^t]$$
 (88)

Solving for  $A_1^{t+\Delta t}$  and  $P_1^{t+\Delta t}$  we get:

$$A_1^{t+\Delta t} = A_1^t + [A_1^0 + (R_1 - R_2) + D_1 + M_1 - A_1]^t \Delta t_A$$
 (89)

$$P_i^{t+\Delta t} = P_i^t + [P_i^0 - D_i - M_i - P_i]^t \Delta t_p$$
 (90)

) where

$$\Delta t_{A} = \frac{A_{T} \Delta t}{V_{A}^{f} \rho_{A}}$$
 (dimensionless)

$$\Delta t_{\mathbf{p}} = \frac{P_{\mathbf{T}} \Delta t}{V(1-f_{\mathbf{A}})\rho_{\mathbf{p}}}$$
 (dimensionless)

Equations analogous to 89 and 90 may now be written for all components in the organic phase (except P) which is given by Equation 63) and for all components in the acid phase, and then solved via the Euler algorithm. This procedure was incorporated into the original computer program which was written to solve the unsteady state equations. Although the procedure is theoretically and numerically consistent, it was found that because of the large numerical values of the diffusion terms, a very small integration step size (At) was required in order to obtain numerical stability and generate convergent solutions. This was not acceptable, since use of an extremely small  $\Delta t$  resulted in intolerably long computer runs. To avoid this difficulty, an alternate solution procedure was devised based on the fact that the accumulation of nitrobody in the acid phase is small compared to that in the organic phase. This allows the integration of the nitrobody components to be carried out over the combined total amount of nitrobody in both phases, while the integration of the acid components in the acid phase is conducted as before. The concentration of the individual nitrobody components in the acid phase can then be obtained by solving the appropriate steady state equations after the integration has been carried out. In effect, it is assumed that the nitrobody in the acid phase is in equilibrium with that in the organic phase. Implementation of the procedure described above is carried out by first adding Equations 84 and 85 to give:

$$P_1^0 + A_1^0 + R_1 - R_3 - (P_1 + A_1) = \frac{V(1-f_A)\rho_P}{P_T} \frac{dP_1}{dt} + \frac{Vf_A\rho_A}{A_T} \frac{dA_1}{dt}$$
(91)

(Note that the troublesome mass transfer terms have dropped out of Equation 81.)

Now, let the total nitrobody in both phases be given by:

$$T_1 = P_1 + A_1 \tag{92}$$

so that

$$P_1 = T_1 - A_1 \tag{93}$$

Substitution of Equations 92 and 93 into Equation 91 yields:

$$P_1^0 + A_1^0 + R_1 - R_3 - T_1 = \frac{V(1-f_A)\rho_P}{P_T} \frac{dT_1}{dt} - \frac{V(1-f_A)\rho_P}{P_T} \frac{dA_1}{dt}$$

$$+\frac{V_{A}^{f}\rho_{A}}{A_{T}}\frac{dA_{1}}{dt}$$
 (94)

$$P_1^0 + A_1^0 + R_1 - R_3 - T_1 = \frac{V(1-f_A)\rho_p}{P_T} \frac{dT_1}{dt} + \frac{Vf_A\rho_A}{A_T}$$

$$-\frac{V(1-f_A)\rho_P}{P_T} \frac{dA_1}{dt}$$
 (95)

Since

$$\frac{d A_1}{dt} <<< \frac{d T_1}{dt}$$

we can write:

$$P_1^0 + A_1^0 + R_1 - R_2 - T_1 \approx \frac{V(1-f_A)\rho_P}{P_T} \frac{dT_1}{dt}$$
 (98)

In terms of discrete quantities:

$$T_1^{t+\Delta t} = T_1^t + \Delta t_p [P_1^0 + A_1^0 + R_1 - R_8 - T_1]^t$$
 (97)

Equation 97 is called the normal or forward difference form of the discrete equation, where the term in brackets is evaluated at the beginning of the interval. However, the efficiency and stability of the numerical integration procedure is enhanced if the backward difference form is used, in which case the bracketed term is evaluated at the end of the interval. In this case:

$$T_1^{t+\Delta t} = T_1^t + \Delta t_p [P_1^0 + A_1^0 + R_1 - R_1 - T_1]^{t+\Delta t}$$
 (98)

Solving for  $T^{t+\Delta t}$ , we get:

$$T_1^{t+\Delta t} = \frac{T_1^t + \Delta t_p \left[ P_1^0 + A_1^0 + R_1 - R_2 \right]^{t+\Delta t}}{1 + \Delta t_p}$$
 (99)

But (from Appendix A),

$$R_3 = A_1 (G_3 \delta + G_{35} \gamma) = A_1 F_3$$
 (100)

From the steady state equation

$$A_1^0 + R_1 - R_2 + D_1 + M_1 = A_1 = 0$$
 (101)

we can determine a value for A<sub>1</sub> as follows:

$$A_1 = A_1^0 + R_1 - G_3 \delta A_1 - G_{35} \gamma A_1 + \eta \left(\frac{P_1}{P_{nb}} - \frac{A_1}{A_{nb}}\right) + M_{nb} \left(\frac{P_1}{P_{NB}} + \frac{A_1}{A_{nb}}\right) (102)$$

$$(1 + G_{3} \delta + G_{3} \delta \gamma + \frac{\eta}{A_{nb}} - \frac{M_{nb}}{A_{nb}}) A_{1} = A_{1}^{C} + R_{1} + P_{1} \left(\frac{\eta}{P_{nb}} + \frac{M_{nb}}{P_{nb}}\right)$$

$$(103)$$

But 
$$P_1 = T_1 - A_1$$
 so that

$$(1 + G_8 \delta + G_{88} \gamma + \frac{\eta}{A_{nb}} - \frac{M_{nb}}{A_{nb}}) A_1 = A_1^0 + R_1$$

$$+ T_1 \left( \frac{\eta}{P_{nb}} + \frac{M_{nb}}{P_{nb}} \right) - A_1 \left( \frac{\eta}{P_{nb}} + \frac{M_{nb}}{P_{nb}} \right)$$
 (104)

$$(1 + G_3 \delta + G_{33} \gamma + \frac{\eta}{A_{nb}} - \frac{M_{nb}}{A_{nb}} + \frac{\eta}{P_{nb}} + \frac{M_{nb}}{P_{nb}}) A_1 = A_1^{\circ}$$

$$+R_1+T_1\left(\frac{\eta}{P_{nb}}+\frac{M_{nb}}{P_{nb}}\right) \tag{105}$$

$$A_{1} = \frac{A_{1}^{0} + R_{1} + T_{1} \left( \frac{\eta}{P_{nb}} + \frac{M_{nb}}{P_{nb}} \right)}{(1 + G_{3}\delta + G_{3}\gamma + \frac{\eta}{A_{nb}} - \frac{M_{nb}}{A_{nb}} + \frac{\eta}{P_{nb}} + \frac{M_{nb}}{P_{nb}})}$$
(106)

$$A_{1} = \frac{A_{1}^{O} + R_{1} + T_{1} (C_{X})}{F_{2} + C_{x}}$$
 (107)

where:

$$C_{X} = (\eta/P_{nb} + M_{nb}/P_{nb})$$

$$C_{g} = (1 + \eta/A_{nb} - M_{nb}/A_{nb} + C_{X})$$

$$F_{g} = G_{g} + G_{gg} \gamma$$

Substitution of Equations 100 and 107 into Equation 99 gives:

$$T_{1}^{t+\Delta t} = \frac{T_{1}^{t} + \Delta t_{p} \left[A_{1}^{0} + P_{1}^{0} + R_{1} - \frac{F_{3}}{F_{3} + C_{z}} \left(A_{1}^{0} + R_{1} + C_{x} T_{1}^{t+\Delta t}\right)\right]^{t+\Delta t}}{1 + \Delta t_{p}}$$
(108)

Rearrangement of Equation 108 yields:

$$T_1^{t+\Delta t} = \frac{T_1^t + \Delta t_p X}{1 + \Delta t_p Y}$$
 (109)

where

$$X = \left[ P_1^{0} + (R_1 + A_1^{0}) \left( 1 - \frac{F_8}{F_8 + C_g} \right) \right]$$
 (110)

$$Y = \left[1 + \left(\frac{F_3}{F_3 + C_2}\right) C_X\right] \tag{111}$$

The value of  $A_2^{\ t+\Delta t}$  can now be computed from Equation 107 using the value of T determined in Equation 109.  $A_{nb}^{\ t}, M_{nb}^{\ t}, \gamma$ , and  $\delta$  in Equation 107 are computed using the values of  $A_1^{\ t}$  and  $P_1^{\ t+\Delta t}$  this has not posed any problems in the execution of the program. Equations similar to 109 can now be written to determine  $T_2^{\ t+\Delta t}$  through  $T_6^{\ t+\Delta t}$  with  $A_2^{\ t+\Delta t}$  through  $A_8^{\ t+\Delta t}$  being calculated from equations analogous to 107.

For the acid components in the acid phase, forward difference integration is employed.

$$A_{\theta}^{t+\Delta t} = A_{\theta}^{t} + [A_{\theta}^{0} + P_{\theta}^{0} - P_{\theta} - R_{1} - R_{1} - R_{2} - R_{3} - R_{4} - R_{6} - R_{6}$$

$$- 13 R_{7} - 12.5 R_{\theta} - 9 R_{10} - 12.5 R_{\theta} G + (.33 + .67 RP) R_{\theta}$$

$$- A_{\theta}]^{t} \Delta t_{A}$$
(112)

$$AL^{t+\Delta t} = AL^{t} + [A_{18}^{O} - A_{14}^{O} + R_{1} + R_{8} + R_{8} + R_{4} + R_{8} + R_{8} + R_{8} + R_{1} + R_{1} + R_{1} + R_{1} + R_{2} + R_{3} + R_{4} + R_{5} + R_{$$

where 
$$AL^{t+\Delta t} = A_{13}^{t+\Delta t} - A_{14}^{t+\Delta t}$$
 (114)

If AL < 0 
$$A_{14} = 0$$

$$A_{14} = AL$$
and if AL  $\geq$  0 
$$A_{14} = AL$$

$$A_{12}^{t+\Delta t} = A_{12}^{t} + [A_{12}^{0} + (11 R_{7} + 14.5 R_{6} + 9 R_{16} + 14.5 R_{6G})] (1-FD)$$

$$- A_{12}^{t} \Delta t_{A} \qquad (115)$$

$$A_{SO}^{t+\Delta t} = A_{SO_4}^t + [A_{11}^O + A_{14}^O - (11 R_T + 14.5 R_B + 9 R_{1B} + 14.5 R_{BG}) (1-F_D) - A_{SO_4}^{t}]^t \Delta t_A$$
(116)

$$A_{11}^{t+\Delta t} = A_{SO_4}^{t+\Delta t} - A_{14}^{t+\Delta t}$$
 (117)

#### Solution Procedure for Dynamic Simulation

The primary sequence of operations which must be executed in order to obtain the unsteady state solution of the nitration section equations is as follows:

- Step 1: Enter the initial steady state of the process. This includes the composition and flow rates of the ideally separated acid and organic phases from each of the 14 process vessels, as well as the corresponding feed rates, compositions, nitrator temperatures, and notch settings.
- Step 2: Process the initial conditions through the physical separation equations for all six separators to get initial conditions on the recycle streams.
- Step 3: Read in the perturbations to the independent variables which constitute the deviations from steady state.
- Step 4: Begin execution of the vessel equations for Vessel 1 (Nitrator 1A) by making up the terms required to compute the generation expressions  $D_1$ ,  $M_1$ , and  $R_2$ .
- Step 5: Integrate using Equations 107 to 117 to get the values of P<sub>1</sub> and A<sub>4</sub> at the end of the first time step.
- Step 6: Store these values as the current outputs from Nitrator 1A, and use them to make up the inputs to Nitrator 1B.
  - Step 7: Repent Steps 4 and 5 for Nitrator 1B.
- Step 8: Store these values as the current outputs from Nitrator 1B and use them to make up the inputs to Separator 1.
  - Step 9: Repeat Steps 4 and 5 for Separator 1.
- Step 10: Store these values as the current ideally separated outputs from Separator 1.
- Step 11: Process the current values of the ideally separated output from Separator 1 through the physical separation equations to give the current composition and flow of the three streams leaving separator 1.

Step 12: Store these values for use in making up inputs to other vessels.

Step 13: Begin execution of vessel equations for Nitrator 2 and continue the procedure for all remaining vessels through Separator 6.

At this point, one complete pass will have been made across the entire nitration reactor train for the first time step with the values of each vessel's output used to make up the necessary inputs for execution during the second time step. This procedure is now repeated for n time steps as desired. The program currently employs a time step of .01 hours (36 seconds). Thus, to simulate 12 hours of process operation, 1200 time steps must be executed. Typically, the execution time required for such a case is about 120 seconds on the CDC 6500 computer giving an average of 0.1 second of machine time per time step.

#### Program Description

The nitration section dynamic simulation consists of a main program, four subroutine subprograms, and one function subprogram. Appendix B presents a complete program listing; Appendix C shows the program flow chart; Appendix D gives the program nomenclature; and Appendix E presents the input data format.

All programs are written in FORTRAN IV for compilation on the CDC FORTRAN extended (FTN) compiler, version 3.0 and require 65K (octal) words on the CDC 6500 computer for complete execution. Word length on the CDC 6500 is 60 bits. Since there are no machine-dependent routines, the program should execute on any comparable computer with only minor modifications. It should be noted that a number of library subroutines (NANCY, NUCHAR, and NANCYM) are used to plot desired output.

The MAIN program reads in all data and parameter values, monitors and controls the sequential execution of the vessel and physical separation equations, and keeps track of all vessel inputs and outputs. The program also computes the values of a number of process performance indicators from the current values the vessel outflows at various times during the run and plots them as a function of time. These performance indicators constitute the primary output of the dynamic simulation and include: concentration of DNT in the crude TNT (wt %), flow rate of crude TNT (lb/hr); concentration of nitrobody in the spent acid (wt %), the fraction DNT in the spent acid nitrobody (wt %), concentration of water in the spent

acid (wt %), flow rate of spent acid (lb/hr), total carbon and nitrogen oxides in the off-gas (lb-moles/hr), heat loads in each nitrator (BTU/hr), concentration of nitric acid in the external recycle from each separator (wt % actual nitric), ratio of nitric to sulfuric acid in each nitrator, the amount of nitration occurring in each nitrator, and the specific gravities of the external acid recycle and organic streams leaving each separator. The values of e' and e' for each separator given by Equations 66 and 67 or each separator given g

Subroutine INPUT is called by the main program to read in the starting values of the independent variables including raw material feed rates and compositions, nitrator temperatures, and internal recycle notch setting, as well as to read in the magnitude and time of the desired perturbations to these quantities. Since feed rate data is entered in lb/hr, the subroutine makes the appropriate conversions to molar units. Also, the exponential factors in the rate equations (e.g.,

$$K_{i} \exp \left[-\frac{F_{i}}{R} \left(\frac{1}{T} - \frac{1}{T_{R}}\right)\right]$$

are precomputed in INPUT in order to increase the efficiency with which the vessel equations are executed.

The heart of the dynamic simulation is subroutine SUB. Here, all of the vessel and physical separation equations are executed and integrated each time the subroutine is called by the main program as it steps through the reactor train. In effect, the main program provides the necessary input data to SUB (i.e. the main program tells SUB whether it is a nitrator or set trator and then which one it is in the reactor train; it provides vessel equations for that vessel such as volume and notch setting; it provides the inject and output phase compositions for the vessel from the previous time step). SUB then generates the output of the vessel and returns. SUB also computes a temperature rise for each separator based on the adiabatic heat generated by the small amount of reaction which takes place there.

Subroutine SUB1 computes the change in the calculated values of the phase compositions for each vessel from one iteration to the next. An iteration is defined as the execution of m time steps where m is specified at the beginning of the run. SUB1 also flags that component in each vessel which shows the maximum change between iterations.

Subroutine SUB2 executes the physical separation equations for each separator using the steady state phase compositions of the input data. SUB2 is called by the main program prior to execution of the first time step only in order to initialize the recycle flows. The physical separation equations are thereasfter executed in subroutine SUB as the run progresses.

The final subprogram in the dynamic simulation is function ENTH. This function is used in conjunction with the nitrator heat balance calculations executed by the main program. Its purpose is to calculate the enthalpy of any specified flow stream in the process by interpolating enthalpy-composition data which exits as a data table within the function. The enthalpies of the input and output streams are combined with the heats of reaction and heats of mixing to compute each nitrator's heat duty.

#### The Steady State Simulation

The steady state simulation is a computer program which, for a given set of nitration section independent variables, will execute the vessel and physical separation equations to give the steady state values of the composition and flow rates (moles/hr) of the exiting streams from each of the 14 process vessels. In addition, the values of a number of process performance functions are computed.

At steady state, the accumulation terms in Equations A-57 to A-82 go to zero so that differential equation system of the dynamic case becomes one of simultaneous algebraic equations. Because of the implicit nature of these equations, and also because of the presence of numerous recycle streams, a multilevel (nested) iteration procedure is used along with a simultaneous equation solving technique to arrive at the final solution.

#### Formulation of Equations

For the nitrobody components in the acid phase, the following system of steady state equations can be written:

For component 1: 
$$A_1^0 - R_3 + D_1 + M_1 - A_1 = 0$$
 (118)

$$A_1 + R_3 - D_1 - M_1 = A_1^0 ag{119}$$

Substitution of Equations 16, 20, 21, and 38 from Appendix A into Equation 119 gives

$$A_1 + (G_3 8 + G_3 S \gamma) A_1 - \eta \left(\frac{P_1}{P_{nb}} - \frac{A_1}{A_{nb}}\right) - M_{nb} \frac{A_1}{A_{nb}} - M_{nb} \frac{P_1}{P_{nb}} = A_1^0$$
(120)

Simplifying,

$$A_1 (1 + G_3 \delta + G_3 S \gamma) - C_1 \left(\frac{P_1}{P_{nb}} - \frac{A_1}{A_{nb}}\right) - C_A A_1 - C_R P_1 = A_1^{\circ}$$
(121)

where:  $C_1 = \eta$ 

$$C_A = \frac{M_{nb}}{A_{nb}}$$
 for  $M_{nb} < 0$   $C_A = 0$  for  $M_{nb} \ge 0$ 

$$C_R = \frac{M_{nb}}{P_{nb}}$$
 for  $M_{nb} \ge 0$   $C_R = 0$  for  $M_{nb} < 0$ 

Further rearrangement of Equation 121 yields:

$$(1 + G_0 \delta + G_0 g \delta + \frac{G_1}{A_{nb}} - G_A) A_1 - \left(\frac{C_1}{P_{nb}} + C_R\right) P_1 = A_1^C$$
 (122)

Similarly, for component 2:

$$A_2^0 - R_4 + D_2 + M_2 - A_2 = 0 ag{123}$$

$$A_2 + R_4 - D_3 - M_2 = A_3^0 ag{124}$$

Appropriate substitution from Appendix A yields:

$$A_1 + (G_48 + G_{4S} \gamma) A_2 - \frac{C_1}{P_{nb}} P_1 + \frac{C_1}{A_{nb}} A_2 - C_A A_3 - C_R P_2 = A_2^O$$
 (125)

$$A_2 (1 + G_4 \delta + G_4 S \gamma + \frac{C_1}{A_{nb}} - C_A) - (\frac{C_1}{P_{nb}} + C_R) P_2 = A_2^0$$
 (126)

For component 3:

$$A_8 + R_8 - R_8 - R_{VA} - R_{0A} - R_{0A} + D_8 + M_8 - A_8 = 0$$
 (127)

$$A_3 - R_3 + R_8 + R_{7A} - R_{8A} + R_{9A} - D_3 - M_8 = A_9^0$$
 (128)

$$A_{3} - (G_{3}8 + G_{3}S \gamma) A_{1} + G_{5}\gamma A_{3} + G_{7}\gamma A_{3} + \frac{VG_{9}f_{A}A_{2}}{Q_{A}^{2}} A_{3} + G_{9}\gamma A_{3}$$

$$-\frac{C_1}{P_{NB}}P_8 + \frac{C_1}{A_{NB}}A_8 - C_AA_8 - C_RP_8 = A_8^0$$
 (129)

$$A_{3}\left(1+G_{8}\gamma+G_{7}\gamma+\frac{VG_{8}f_{A}A_{9}}{Q_{A}^{3}}+G_{9}\gamma+\frac{C_{1}}{A_{nb}}-C_{A}\right)-\left(\frac{C_{1}}{P_{nb}}+C_{R}\right)P_{3}$$

$$= A_3^{C} + (G_3 \delta + G_{3S} \gamma) A_1$$
 (130)

For component 4:

$$A_4^0 + R_4 - R_6 - R_{7M} - R_{8M} - R_{9M} + D_4 + M_4 - A_4 = 0$$
 (131)

$$A_4 - R_4 + R_6 + R_{\uparrow M} + R_{\delta M} + R_{\delta M} - D_4 - M_4 = A_4^0$$
 (132)

$$A_4 - (G_4 B + G_{48} \gamma) A_2 + G_8 \gamma A_4 + \frac{VG_0 f_A A_0}{Q_A^2} A_4 + G_0 \gamma A_4$$

$$-\frac{C_1}{P_{nh}}P_4 + \frac{C_1}{A_{nh}}A_4 - C_AA_4 - C_RP_4 = A_4^0$$
 (133)

$$A_{4} \left(1 + G_{0}\gamma + G_{7}\gamma + \frac{VG_{0}f_{A}A_{0}}{Q_{A}^{2}} + G_{0}\gamma + \frac{C_{1}}{A_{nb}} - C_{A}\right)$$

$$-\left(\frac{C_1}{P_{nb}} + C_R\right) P_4 = A_4^0 + (G_4 \delta + G_4 S \gamma) A_2$$
 (134)

For component 5:

$$A_8^0 + R_8 - R_{10A} + D_8 + M_8 - A_8 = 0$$
 (135)

$$A_8 - R_8 + R_{10A} - D_8 - M_8 = A_8^0$$
 (136)

$$A_8 - G_8 \gamma A_8 + \frac{VG_{10}f_A A_{17}}{Q_A^8} A_8 - \frac{C_1}{P_{nb}} P_8 + \frac{C_1}{A_{nb}} A_8 - C_A A_8$$

$$-C_{\mathbf{R}}P_{\mathbf{k}} = A_{\mathbf{k}}^{\mathbf{O}} \tag{137}$$

$$A_{8} \left(1 + \frac{VG_{10}f_{A}A_{17}}{Q_{A}^{8}} + \frac{C_{1}}{A_{nb}} - C_{A}\right) - \left(\frac{C_{1}}{P_{nb}} + C_{R}\right)P_{8}$$

$$= A_{8}^{O} - G_{8}\gamma A_{8}$$
(138)

For component 6:

$$A_6^0 + R_6 - R_{10M} + D_6 + M_6 - A_6 = 0$$
 (139)

$$A_6 - R_6 + R_{10M} - D_6 - M_6 = A_6^0$$
 (140)

$$A_6 - G_6 \gamma A_4 + \frac{VG_{16}f_A^{}A_{17}}{Q_A^2} A_6 - \frac{C_1}{\frac{P}{nb}} P_6 + \frac{C_1}{\frac{A}{nb}} A_6 \cdot C_A^{}A_6$$

$$-C_{R}P_{6}=A_{6}^{O} \qquad (141)$$

$$A_{6} \left(1 + \frac{VG_{16}f_{A}A_{17}}{Q_{A}^{2}} + \frac{C_{1}}{A_{nb}} - C_{A}\right) - \left(\frac{C_{1}}{P_{nb}} - C_{R}\right) P_{6}$$

$$= A_{6}^{0} + G_{6}\gamma A_{4} \qquad (142)$$

For component 7:

$$A_{\tau}^{0} + R_{10} + D_{\tau} + M_{\tau} - A_{\tau} = 0 ag{143}$$

$$A_7 - R_{10} - D_7 - M_7 = A_7^0 ag{144}$$

$$A_{7} - \frac{VG_{10}f_{A}A_{17}}{Q_{A}^{2}} \quad (A_{8} + A_{4}) - \frac{C_{1}}{P_{nb}} \quad P_{7} + \frac{C_{1}}{A_{nb}} \quad A_{7} - C_{A}A_{7}$$
$$- C_{R}P_{7} = A_{7}^{0} \qquad (145)$$

$$A_{1} \left(1 + \frac{C_{1}}{A_{nb}} - C_{A}\right) - \left(\frac{C_{1}}{P_{nb}} + C_{R}\right) P_{1} = A_{1}^{0}$$

$$+ \frac{VG_{10}f_{A}A_{17}}{Q_{A}^{2}} (A_{0} + A_{0}) \qquad (146)$$

For component 8:

$$A_8^0 + 3 R_{10} + D_0 + M_0 - A_0 = 0 (147)$$

$$A_8 - 3 R_{10} - D_8 - M_8 = A_8^0$$
 (148)

$$A_{0} = \frac{3VG_{10}f_{A}A_{17}}{Q_{A}^{2}} (A_{0} + A_{0}) = \frac{C_{1}}{P_{nb}} P_{0} + \frac{C_{1}}{A_{nb}} A_{0} = C_{A}A_{0}$$
$$= -C_{R}P_{0} = A_{0}^{O}$$
(149)

$$A_{e} \left(1 + \frac{C_{1}}{A_{hb}} - C_{A}\right) - \left(\frac{C_{1}}{P_{hb}} + C_{R}\right) P_{e} = A_{e}^{0} + \frac{3VG_{1e}f_{A}A_{1f}}{Q_{A}^{2}} (A_{e} + A_{e})$$
(150)

For the nitrobody species in the organic phase, the steady state vessel equations can be written as follows:

$$P_1^0 + R_1 - M_1 - D_1 - P_1 = 0 (151)$$

$$P_1 - R_1 + M_1 + D_1 = P_1^0 ag{152}$$

$$P_1 - D_{10} (1 - G_1) + C_A A_1 + C_R P_1 + \frac{C_1}{P_{nb}} P_1 - \frac{C_1}{A_{nb}} A_1 = P_1^0$$
 (153)

but 
$$D_{10} = \eta_{Tol} \frac{P_{10}}{P_{nb}} = C_1^{'} P_{10}$$
 (154)

so that,

$$P_1 - C_1^{\dagger} P_{10} (1-G_1) + A_1 (C_A - \frac{C_1}{A_{nb}}) + P_1 (C_R + \frac{C_1}{P_{nb}}) = P_1^{\circ}$$
(155)

$$(C_A - \frac{C_1}{A_{nb}}) A_1 + (1 + C_R + \frac{C_1}{P_{nb}}) P_1 = P_1^0$$

$$+ C_1' P_{10} (1 - G_1)$$
 (156)

For component 2:

$$P_8^O + R_8 - M_2 - D_2 - P_3 = 0$$
 (157)

$$P_2 - R_2 + M_2 + D_2 = P_3^0 ag{158}$$

$$P_{8} - C_{1}^{1} P_{10} G_{1} + C_{A} A_{3} + C_{R} P_{8} + \frac{C_{1}}{P_{nb}} P_{8} - \frac{C_{1}}{A_{nb}} A_{8} = P_{8}^{O}$$
 (159)

$$(C_A - \frac{C_1}{A_{nb}}) A_2 + (1 + C_R + \frac{C_1}{P_{nb}}) P_3 = P_3^0 + C_1^{\dagger} P_{10} G_1$$
 (160)

For component 3:

$$P_{a}^{O} - M_{a} - D_{a} - P_{a} = 0 (181)$$

$$P_3 + M_3 + D_3 = P_3^0$$
 (162)

$$P_{8} + C_{A}A_{8} + C_{R}P_{8} + \frac{C_{1}}{P_{nb}}P_{8} - \frac{C_{1}}{A_{nb}}A_{8} = P_{8}^{0}$$
 (163)

$$(C_A - \frac{C_1}{A_{nb}}) A_1 + (1 + C_R + \frac{C_1}{P_{nb}}) P_2 = P_2^0$$
 (184)

For component 4:

$$P_4^0 - M_4 - D_4 - P_4 = 0 (185)$$

$$P_4 + M_4 + D_4 = P_4^0 ag{166}$$

$$(C_A - \frac{C_1}{A_{nb}}) A_4 + (1 + C_R + \frac{C_1}{P_{nb}}) P_4 = P_4^0$$
 (167)

Similarly, for components 5 through 8:

$$C_{21}A_{6} + C_{22}P_{6} \approx P_{6}^{O} \tag{188}$$

$$C_{11}A_{0} + C_{12}P_{0} = P_{0}^{O} \tag{169}$$

$$C_{31}A_7 + C_{33}P_7 = P_7^{O} (170)$$

$$C_{21}A_{0} + C_{22}P_{0} = P_{0}^{O} (171)$$

where:

$$C_{11} = C_A - \frac{C_1}{A_{nb}}$$
 and  $C_{11} = (1 + C_R + \frac{C_1}{P_{nb}})$ 

Now, for toluene:

$$P_{10}^{0} - D_{10} P_{10} = 0 (172)$$

$$P_{10} + D_{10} = P_{10}^{O} \tag{173}$$

$$P_{10} (1 + C_1^{'}) = P_{10}^{O}$$
 (174)

$$P_{10} = P_{10}^{0}/(1+C_{1}^{1}) \tag{175}$$

For the acid species in the acid phase, the steady state vessel equations are written as follows:

$$A_0^0 + P_0^0 - P_0 - D_{10} - R_0 - R_0 - R_0 - R_0 - 13R_7 - 12.5R_0$$

$$-9R_{10} - 12.5R_0G + (.33 + .67 RP) R_0 - A_0 = 0$$
 (176)

Making the appropriate substitutions from Appendix A gives:

$$A_{\theta} + P_{\theta} + C_{1}^{'} P_{1\theta} + (G_{1}B_{+}G_{2}g\gamma) A_{1} + (G_{4}B + G_{4}g\gamma)A_{2} + G_{5}\gamma A_{2}$$

$$+ G_{6}\gamma A_{4} + 13G_{7}\gamma (A_{8} + A_{4}) + 12.5 \left(\frac{VG_{6}f_{A}}{Q_{A}} - [HNO_{3}]\right) (A_{2} + A_{4})$$

$$+ 12.5G_{6}\gamma (A_{2} + A_{4}) + 9 \left(\frac{VG_{10}f_{A}}{Q_{A}} - [NO_{8}^{+}]\right) (A_{6} + A_{6})$$

$$- (.33 + .67RP)F_{D}C_{9} = A_{8}^{O} + P_{9}^{O}$$

$$A_{0} + P_{0} + C_{1}^{'}P_{10} + F_{2}A_{1} - F_{4}A_{2} + \left(G_{5}\gamma + 13G_{7}\gamma + 12.5\frac{VG_{9}f_{A}}{Q_{A}} - [HNO_{3}]\right)$$

$$+ 12.5G_{9}\gamma A_{3} + \left(G_{6}\gamma + 13G_{7}\gamma + 12.5\frac{VG_{9}f_{A}}{Q_{A}} - [HNO_{8}] + 12.5G_{9}\gamma\right) A_{4}$$

$$+ 9 \frac{VG_{10}f_{A}}{Q_{A}} - [NO_{3}^{+}] A_{3} + 9 \frac{VG_{10}f_{A}}{Q_{A}} - [NO_{8}^{+}] A_{6} - (.33 + .67RP)F_{D}G_{9}$$

$$= A_{9}^{O} + P_{9}^{O}$$

$$(177)$$

Also, 
$$P_0 = \left(\frac{1.51 A_{18} P_T}{A_T A_{11}}\right) A_0$$
 (178)

$$A_{11}^{O} + A_{14}^{O} - C_{bp} - A_{8O_{4}} = 0$$
 (179)

$$A_{SO_4} + (1 - F_D)C_3 = A_{11}^O + A_{14}^O$$
 (180)

$$A_{18}^{O} + C_{9}P - A_{12} = 0 (181)$$

$$A_{18} - C_{0P} + A_{18}$$
 (182)

$$A_{18}^{O} - A_{14}^{O} + R_{1} + R_{2} + R_{3} + R_{4} + R_{5} + R_{5} + 15R_{7} + 16.5R_{8} + 12R_{10}$$

$$+ 16.5R_{9Q} - (.33R_{p} + .67)R_{9} - A_{1} = 0$$
(183)

$$A_{L} - C_{1}^{\dagger} P_{10} - F_{3}A_{1} - F_{4}A_{3} - \left(G_{6}\gamma + 15G_{1} - 16.5 \frac{VG_{0}f_{A}}{Q_{A}} [HNO_{3}]\right)$$

$$+ 16.5G_{0}\gamma A_{3} - \left(G_{0}\gamma + 18G_{\gamma}\gamma + 18.5 \frac{VG_{0}f_{A}}{Q_{A}} [HNO_{3}]\right)$$

$$+ 16.5G_{0}\gamma A_{4} - 12 \frac{VG_{10}f_{A}}{Q_{A}} [NO_{3}^{+}] A_{4} - 12 \frac{VG_{10}f_{A}}{Q_{A}} [NO_{3}^{+}] A_{6}$$

$$+ (.33RP + .87) F_{D}C_{3} = A_{13}^{O} + A_{14}^{O}$$

$$(184)$$

Equations 122, 128, 130, 134, 138, 142, 146, 150, 156, 160, 164, 167, 168, 169, 170, 171, 175, 177, 178, 179, 182, and 184 are linear in the following unknowns:  $A_1$  through  $A_2$ ,  $A_{SO_4}$ ,  $A_{12}$ ,  $A_L$ , and  $P_1$  through  $P_{10}$ . This

constitutes a system of 22 equations and 22 unknowns which is solvable provided the coefficients of all the unknown terms can be determined.

Solution Procedure for the Steady State Simulation

Execution of the steady state simulation consists mainly of the simultaneous solution of the 22 vessel equations for each of the 14 vessels in the nitration train. Although these equations could be solved simultaneously via Gauss reduction, it was decided that because of the sparcity of the coefficient matrix, a straightforward direct substitution approach would be more efficient. This will become clear as the step-by-step solution procedure is outlined below. The physical separation equations must also be employed here as in the dynamic case to convert computed pure phase outputs to real stream outputs. As mentioned previously, a multilevel or nested iterative procedure is used because of (1) the implicit nature of the vessel equations (the coefficients of the linear unknowns are functions of these unknowns), and (2) the numerous recycle streams in the process.

The primary sequence of operations which must be executed in order to obtain the steady solution is as follows:

Step 1: Enter an assumed solution for the phase outputs from each of the 14 process vessels.

Step 2: Enter the values of the process's independent variables for which the steady state solution is desired.

Step 3: Using the assumed solution for the vessel phase outputs, compute the flow rates and compositions of all recycle streams in the process via the physical separation equations.

Step 4: Begin the iterative solution of the vessel equations for the first vessel (Nitrator #1A). Start by computing the values of all coefficients of the  $22 \times 22$  vessel equation matrix from the assumed solution for that vessel. Also, make up the correct phase inputs to Nitrator 1A.

Step 5: Solve Equation 175 for P<sub>10</sub>.

Step 6: Solve Equations 122 and 156 simultaneously for values of  $A_1$  and  $\,P_1$  .

Step 7: Solve Equations 128 and 180 simultaneously for values of  $A_2$  and  $P_2$ .

Step 8: Solve Equations 130 and 184 simultaneously for values of  $A_a$  and  $P_a$ .

Step 9: Solve Equations 134 and 167 simultaneously for values of  $A_4$  and  $P_4$ .

Step 10: Solve Equations 138 and 168 simultaneously for values of  $A_{\delta}$  and  $P_{\delta}$  .

Step 11: Solve Equations 142 and 169 simultaneously for values of  $A_{\alpha}$  and  $P_{\alpha}$ .

Step 12: Solve Equations 146 and 170 simultaneously for values of  $A_7$  and  $P_7$ .

Step 13: Solve Equations 150 and 171 simultaneously for values of  $A_{\alpha}$  and  $P_{\alpha}$ .

Step 14: Solve Equations 178 and 178 simultaneously for values of  $A_{\theta}$  and  $P_{\theta}$  .

Step 15: Solve Equation 182 for A11.

Step 16: Solve Equation 184 for A<sub>T</sub>.

Step 17: Use the logic of Equations A-70 and A-71 to determine values for  $A_{14}$  and  $A_{14}$ .

Step 18: If computed values and assumed values differ by more than a predetermined amount, repeat Steps 5 through 17 using newly computed values for the  $A_i$ 's and  $P_i$ 's.

Step 19: Repeat Step 18 until convergence is achieved on Nitrator 1A.

Step 20: Using the converged solution for Nitrator 1A, and the previously assumed recycle stream flows, make up the correct phase inputs to Nitrator 1B and then repeat Step 4 for Nitrator 1B.

Step 21: Repeat Steps 5 through 17 for Nitrator 1B.

Step 22: Repeat Step 18 for Nitrator 1B.

Step 23: Repeat Step 22 until convergence is achieved on Nitrator 1B.

Step 24: Using the converged solution for Nitrator 1B as the input to Separator 1 and the initially assumed phase outputs, or scute Steps 4 through 19 for Separator 1.

Step 25: Process the converged outputs for Separator 1 through the physical separation equations to get the flow rates and compositions of the recycle streams leaving that separator.

Step 26: Compare the calculated recycle stream between Separator 1 and Nitrator 1B (internal recycle) to that assumed when the inputs to Nitrator 1B were made up. If they differ by a preset amount, use the newly calculated recycle stream, and the previously converged values of the outputs from Nitrators 1A and 1B to repeat Steps 4 through 19 for Nitrator 1B.

Step 27: Repeat Steps 24 through 26 until convergence on the internal recycle stream is achieved.

Step 28: Using the converged Separator 1 outputs and the originally assumed Nitrator 2 output and Separator 3 recycle stream values, execute Steps 4 through 19 for Nitrator 2.

Step 29: Repeat Steps 24 through 27 for Separator 2.

Step 30: Repeat Steps 28 and 29 for Stages 3 through 6 until convergence on all internal recycle streams is achieved. Note, although a converged solution for each stage will have been obtained at this point, no attempt at linking the six stages together through their external recycle streams has yet been made. This must now be done in order to obtain the complete steady state solution for the 14-vessel reactor train.

Step 31: Begin with the external recycle between Separator 6 and Nitrator 5. Using the previously converged outputs from Separators 4, 5, and 6 as Nitrator 5 inputs and the previously converged Nitrator 5 output, execute Steps 4 through 19 for Nitrator 5.

Step 32: Repeat Steps 24 through 27 for Separator 5.

Step 33: Execute Steps 4 through 19 for Nitrator 6 using the newly converged Separator 5 output and previously converged Separator 6 and Nitrator 6 outputs.

Step 34: Repeat Steps 24 through 27 for Separator 6.

Step 35: Compare the newly calculated external acid recycle stream from Separator 6 to that employed in Step 31. If they differ by a preset amount, use the newly calculated value as well as the most recently converged values for the other required flows to repeat Steps 31 through 34.

Step 36: Repeat Step 35 until convergence on the external recycle from Separator 6 is achieved.

Step 37: Continue with this stagewise iteration procedure until the remaining external recycle streams (and thus the entire nitration reactor train) have been converged.

Typically, a steady state run requires approximately 5000 iterations of the vessel equations with convergence limits set at .05 mole/hr for overall vessel composition, .02 moles/hr for components in the internal recycle, and .02 moles/hr for components in the external recycle. Under these conditions, program execution time is approximately 30 seconds on the CDC 8500. Obviously, if tighter convergence is desired, run time increases significantly.

### Program Description

The nitration section steady state simulation consists of a main program, two subroutine subprograms, and one function subprogram. A complete steady state run including the program listing is presented in Appendix F. Appendix G shows the program flow chart; Appendix H gives the program nomenclature; and Appendix I illustrates the input data format.

All programs are written in FORTRAN IV for compilation on the CDC FORTRAN extended (FTN) compiler, version 3.0, and will compile and execute in 55,000 (octal) words on the CDC 6500. Since there are no machine-dependent routines, the program should execute on any comparable computer with only minor modifications. A brief description of the main program and the three subprograms will now be given.

The main program (TNTSIM) reads in all data and parameter values which form the fixed data base for the steady state simulation. Included in this data base is an initial guess at the steady state solution. Also, the program is responsible for reading the particular set of values for the independent process input variables for which the steady state output is desired. All feed streams which are entered as lb/hr are converted to moles/ hr and the Arrhenius factors are precomputed for later use in the rate expressions as are values for eao and eoa . However, the primary function of the main program is to monitor and control the execution of the threelevel convergence procedure employed to arrive at the steady state solution. The program is actually responsible for external recycle convergence and calls the appropriate subroutines for internal recycle or vessel convergence. The program also assigns and updates vessel identification parameters so that it can correctly sequence through the reactor system. After overall convergence is achieved, the main program prints out the steady state values of the ideal phase and actual stream compositions and flow rates from each nitrator and separator in the process (including the off-gas streams) and then proceeds to compute and print out the values of a number of process performance indicators including vessel time constants (hours), overall molar yield, flow rate of crude TNT (lb/hr), DNT in the crude TNT (wt %), raw material cost (\$/lb TNT), spent acid flow (lb/hr) and composition. extent of nitration in each stage, acid composition in each stage (in units similar to those generated by plant operators during control checks), nitrobody density in each stage (lb/ft<sup>2</sup>), and heat duty in each nitrator. An elemental material balance is also run to check the consistency of the converged solution. The complete steady state printout is shown in Appendix F.

Subroutine NITSEP is employed to carry out the internal recycle convergence procedure. It is called by the main program during individual stage convergence and then again during total process convergence. The subroutine compares the current and previously computed values for the internal acid recycle leaving a given separator and does the required updating if the difference exceeds a predetermined value.

The heart of the steady state simulation program is subroutine VESSEL. Here, all of the vessel and physical separation equations are solved to give the output of each nitrator and separator during execution of the nested iteration convergence procedure. The subroutine is thus responsible for the execution and convergence of the primary iteration loop, i.e. the individual vessel. VESSEL is called by the main program and by subroutine NITSEP during the uncoupled stage convergence

procedure and then again by subroutine NITSEP during total process convergence. The calling program provides the necessary input data to VESSEL, such as whether it is to simulate a nitrator or a separator, the values of all input streams and the currently assumed output of the vessel. VESSEL will then execute Steps 4 through 19 of the solution procedure and, if the vessel has been flagged as a separator, process the converged vessel output through the physical separation equations. A modified direct substitution procedure is used, whereby a percentage of the difference between quantities calculated during successive iterations is added to the newly calculated quantity.

The final subprogram in the steady state simulation is function ENTH. It serves the same purpose here as it did in the dynamic simulation.

### Parameter Fitting

As has been illustrated, the vessel equations are interspersed with a number of constants. These include terms associated with kinetic phenomena such as frequency factors, activation energies, and concentration exponents, as well as mass transfer coefficients, equilibrium constants, and solubility factors. In most cases, the appearance of these constants in the vessel equations is theoretically consistent and not simply a function of empirical supposition. An attempt was made to use published, experimentally determined values of constants when such values were available. However, because many needed values were simply not available, and, because most of those that were had been developed under laboratory conditions not comparable to the state of the actual nitration process, it was necessary to fit the constants (parameters) to steady state process operating data (steady state snapshots).

The parameter fitting procedure consisted of (1) assuming an initial value for each of the 27 parameters, (2) executing the steady state simulation with these par meter values and input variables values corresponding to specific data snapshots, (3) comparing computed outputs (molar component flows of all process streams) with observed snapshot values, and (4) making appropriate changes in parameter values to give better agreement between calculated and observed quantities.

A total of 14 steady state snapshots were available for parameter fitting. This data was obtained during normal operations at the production facility located at the Radford Army Ammunition Plant. A typical snapshot is shown in Table I of Appendix J. Of the 14 snapshots, seven were used for fitting with the remaining sets being employed to check the validity of the final fit.

In order to use the snapshot data (which is essentially stream compositions in weight %) for fitting purposes, it was first necessary to convert from units of composition to units of molar flow rate for each component. This was accomplished by processing the snapshot data through a material balance program, the details of which can be found in Reference 1. With the snapshot data in correct form for comparison with the individual stage outputs calculated by the simulation, it was simply required to make repetitive steady state runs and after each run, to evaluate an objective function. This function was in effect a measure of the total weight deviation from observed conditions in all nitration stages. Changes were then made to the parameter set which drove the objective function toward a minimum.

Minimisation of the objective function was accomplished systematically by using a pattern search algorithm which was added to the steady state simulation as a subroutine and controlled program execution. The pattern search technique will be described in detail in the next section of the report when steady state optimization is discussed. A list of parameters with their final fit values is included in Table II of Appendix J.

## APPLICATION OF THE SIMULATION

The computer simulation of the nitration section has been used to carry out a variety of studies on the nitration process. Most significant among these has been the use of the steady state simulation to develop optimized operating conditions and the development of a multivariable control procedure for vessel acid composition using the dynamic simulation to generate the process transfer function.

It is not intended that this report serve as the documentary record of the exploitation of the nitration section simulation. Detailed technical reports on specific studies using the nitration section simulation have been prepared (see Ref 15 to 17). However, it is desired in this volume to give the reader some feeling for the way in which the nitration section simulation can be used. With this in mind, the following description of two major areas of exploitation is presented.

# Steady State Applications

Steady state optimization was carried out by making multiple steady state runs under the control of the same pattern search algorithm that was used during parameter fitting. The pattern search method is one of a number of direct climing (Ref 13) techniques which will move toward an extremum of a multidimensional objective function based on previous information

about the response surface. The particular pattern search algorithm used was an adaptation of a procedure developed by Hooke and Jeeves (Ref 14) which employs accelerated climing and has ridge-following properties.

O

For each steady state run made under control of the pattern search subroutine, an objective function, defined as the raw material cost per pound of TNT, was evaluated. The independent variables over which the search was made included feed composition, feed flow rate, nitrator temperature and internal recycle rate. The feed rate of toluene to the process (and therefore the production rate) was not varied during steady state optimization so that what was sought was the set of independent variables which gave the minimum raw material cost per pound of  $\alpha$ -TNT produced at a fixed production rate. The fixed toluene rate employed during optimization was 2350 lb/hr, which corresponds to a TNT production rate of approximately 57 tons/day based on an 87%  $\alpha$ -yield. The raw material cost per pound of TNT was calculated from the equation:

$$C = \frac{(TxC_T) + (SxC_S) + (NxC_N) + (WxC_W)}{(TxY)}$$
(185)

where: C = raw material cost (\$/lb TNT)

T = toluene flow rate (lb/hr)

S = oleum flow rate (lb/hr)

N = strong nitric acid flow rate (lb/hr)

W = weak nitric acid flow rate (lb/hr)

C<sub>T</sub> = cost of toluene (\$/1b toluene)

 $C_{S} = cost of oleum ($/1b)$ 

C<sub>N</sub> = cost of strong nitric soid (98.5% HNO<sub>2</sub>) (\$/lb)

 $C_w = cost of weak nitric acid (61.0% HNO<sub>2</sub>) ($/1b)$ 

Y = yield (lb TNT/lb toluene)

Values of cost factor employed were based on data supplied by Radford Army Ammunition Plant and thus reflected that plant's raw material cost as of July 1972. Specific values used were:  $C_{\rm T} = .0293$ ,  $C_{\rm S} = .01192$ ,  $C_{\rm W} = .01061$ ,  $C_{\rm N} = .01714$ .

Rather than consider all independent variables simultaneously, a sensitivity analysis was carried out which enabled ranking these variables in the order in which perturbations in them affected the objective function. Only main effect sensitivities were determined. Although the possibility of significant higher order effects could not be discounted, it was felt that the complexity of multiple-effect analysis (including increased machine time requirements) was not justified.

The independent variables were thus divided into five blocks according to their effects on the objective function, and pattern searches were conducted separately over each block. Since the number of variables per search was reduced by this procedure, the pattern search algorithm functioned more efficiently and saved computer time. The complete list of independent variables, ranked in decreasing order of their effects on the objective function and broken down into their search blocks, is shown in Table I of Appendix K.

Because of physical limitations in the actual process, a number of constraints on the independent variables had to be included in the objective function to insure that only feasible sets of process operating conditions were considered. This was accomplished by assigning penalty values to the objective function whenever a constraint value of one or more of the independent variables was under consideration of the search routine. This "brute torce" method worked satisfactorily in keeping the search within feasible areas of the response surface. The constraints employed were based on information supplied by plant operating personnel at Radford and are listed in Table II of Appendix K.

As the optimization proceeded, it became evident that a substantial reduction in cleum was being made by the search program. Because of the magnitude of the predicted reduction (nearly 50% of the baseline feed rate), the validity of the model in a region so far from conditions under which parameters were fit came into question. Also, even if the simulation's predictions were correct, it was felt that plant operating personnel would never agree to such a drastic reduction in cleum flow. But the qualitative result was clear—cut back on cleum flow.

Consequently, the oleum flow rate was treated as a parameter, with sets of optimum conditions being developed for cleum flow rates of 11,000, 10,000, 9,000, 8,000, and 7,000 lb/hr. Of course, the lowest unit cost of TNT is predicted for the lowest cleum flow rate. Predicted optimum conditions for 10,000 lb/hr cleum are shown in Table K3 of Appendix K. For the interested reader, a more complete description of certain aspects of the steady state optimization studies can be found in Reference 3. In general, however, steady state optimization studies have resulted in the development of operating conditions which predict savings on the order of 20% in the raw material cost per pound of TNT.

## Dynamic Applications

The primary application of the dynamic simulation has been its use in conjunction with the design of a supervisory control strategy for acid composition in each stage of nitration. The need to control the composition of the acid phase in each nitration stage is a result of the critical effect which this variable has on the extent of reaction. The object of the control study was thus to develop a procedure whereby predicted optimum acid concentrations could be maintained throughout the process in view of uncontrollable fluctuations in such things as raw material acid composition, internal flows, mixing characteristics, and heat transfer coefficients, as well as step changes in production rate.

It was determined that the way to accomplish this was to control specific indicators of acid concentration in all six external recycle streams and in the emulsion stream flowing from Nitrator 3A to Nitrator 3B by manipulating the seven primary nitric acid feeds and the yellow waterfeeds to the process. A system of nine manipulated and nine controlled variables results. The specific indicator of acid composition is optional and can be either total acidity, actual nitric acid, or nitric to sulfuric acid ratio. Control strategies employing various combinations of these controlled variables have been tested and shown to be feasible and the most promising approach appears to be control of to al acidity and actual nitric acid in Stages 1 and 2 with the nitric to sulfuric ratio being controlled at the remaing five locations. Controlled variables with their proposed manipulated variables are listed in Figure 10.

The details of control strategy development have been documented elsewhere (Ref 15), although a number of significant points should be noted here. Because of the many recycle streams in the process, it was necessary

to develop a decoupling mechanism in order to counteract the disturbances created in adjacent nitrators when a feed change is made. Secondly, because of difficulties in obtaining real time measurements of the controlled variables, it was also necessary to develop a computational scheme to estimate the values of the desired controlled variables from measurable data and certain assumed constant values generated by the steady state program. A block diagram of the overall scheme for acid concentration control is shown in Figure 11. Note that the process simulation provides the necessary process dynamics in the same way, as an assumed transfer function (e.g., a first order lag plus dead time) would have, had not a detailed process model been used.

A number of runs were made with the simulated control scheme in order to develop reasonable values for controller tuning parameters. The open and closed loop responses of the system to a 10% step change in oleum flow are shown in Figures 12 and 13, respectively, for a typical run.

The dynamic simulation was also used in conjunction with a simulation of the nitrator temperature cascade control loop which is included in the software package provided by the Foxboro Company as part of the Volunteer AAP direct digital computer control system, scheduled to begin operation in late 1974. Performance of the temperature control system was tested for normal continuous operation on a single nitrator simulation and also for start-up operations on the complete six-stage nitration section simulation. The details and significant results of this effort can be found in Reference 16.

#### CONCLUSION

The modeling and simulation study of the continuous TNT process has resulted in much being learned about the chemical and physical phenomena which govern the process's behavior. Quantification of process interactions has made possible the generation of optimum operating conditions, improvements in the process design, and has allowed the development of a supervisory control strategy. Implementation of the results of the simulation effort is anticipated during calendar year 1974 when new computer-controlled TNT lines will be started up at Volunteer Army Ammunition Plant in Chattanooga, Tennessee. Only after verification of model predictions on the actual process and use of the model-developed process control strategies, will the full benefit of the simulation undertaking be realized. While simulation studies on other processes are planned, much will depend on the successful implementation of the TNT process simulation study's results.

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APPENDIX A

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**Vessel Equations** 

The following is an ordered set of equations which make up the kinetics and mass transfer model for any vessel in the nitration section of the continuous TNT process. The order in which they are listed represents the logical sequence of calculation employed in SUBROUTINE SUB of the dynamic simulation and SUBROUTINE VESSEL of the steady state simulation program.

# Component Index Assignments

Component	Index (i)
aMNT	1
mMNT	2
ØDNT	3
mDNT	4
αTNT	5
mTNT	6
TNB	7
TNBX	8
HNO <sub>2</sub>	9
Toluene	10
H <sub>2</sub> 8O <sub>4</sub>	11
HNOSO4	12
H <sub>2</sub> O	13
SO <sub>3</sub>	14
Not used	15
Not used	16
NO <sub>3</sub> +	17

Equations

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$$A_{NB} = \sum_{i=1}^{8} A_i \tag{A-1}$$

$$P_{NB} = \sum_{i=1}^{8} P_i + P_{10}$$
 (A-2)

$$A_{T} = A_{NB} = A_{0} + A_{11} + A_{12} + A_{13} + A_{14}$$
 (A-3)

$$P_{T} = P_{NB} + P_{\theta} \qquad (A-4)$$

$$Q_{A} = \sum_{i=1}^{14} \frac{A_{i}}{\rho_{i}}$$
 (A-5)

$$Q_{\mathbf{p}} = \sum_{i=1}^{10} \frac{\mathbf{p}_i}{\mathbf{p}_i} \tag{A-6}$$

$$F_A = \frac{Q_A}{Q_A + Q_P} \tag{A-7}$$

$$F_{\mathbf{p}} = 1 - F_{\mathbf{A}} \tag{A-8}$$

When vessel is a separator,  $F_A = .755$  (A-8a)

$$X_{i} = \frac{A_{i}}{A_{T}} \tag{A-9}$$

$$x_{eq}^{A} = 10^{P} \tag{A-10}$$

Where:

$$P = 3.62 + 0.01023 T - 3.808 X_{18} + 7.718$$

$$(X_1 + X_8) - 8.452 (X_8 + X_8) - 8.292 X_9$$
(A-11)

$$\eta_{A} = VF_{p} \text{ kma} \qquad (A-12)$$

$$\eta_{\mathbf{p}} = VF_{\mathbf{p}} (kma)_{\mathbf{p}}$$
 (A-13)

$$\eta_{Tol} = VF_{P} (kma)_{Tol}$$
 (A-14)

$$\eta = \left(\frac{1}{\frac{1}{\eta_{\mathbf{p}}} + \frac{1}{\eta_{\mathbf{A}} X_{\mathbf{eq}}^{\mathbf{A}}}}\right) \tag{A-15}$$

$$D_{i} = \eta \left(\frac{P_{i}}{P_{NB}} - \frac{A_{i}}{A_{NB}}\right) \quad i = 1.8 \quad (A-16)$$

$$D_{10} = \eta_{Tol} \left( \frac{P_{10}}{P_{NB}} \right)$$
 (A-17)

$$A_{NB} = (X_{eq}^{A}) A_{T}$$
 (A-18)

$$M_{NB} = A_{NB} - A_{NB}^{O} - \sum_{i=1}^{8} D_{i}$$
 (A-19)

If 
$$M_{NB} < 3$$
:  $M_i = M_{NB} \left( \frac{A_i}{A_{NB}} \right)$   $i = 1.8$  (A-20)

If 
$$M_{NB} \ge 0$$
:  $M_i = M_{NB} \left( \frac{P_i}{P_{NB}} \right)$  (A-21)

$$\gamma = \frac{Vf_A}{Q_A} [NO_1^+] \{ k_a [HSO_4^-] + k_b [H_1SO_4] \}$$

$$+ k_{C} [HS_{2}O_{7}^{-}]$$
 (A-22)

or

$$\gamma = k_a \frac{Vf_A}{Q_A} [NO_1^+]$$
 (A-23)

All concentrations are equilibrium values resulting from dissociations according to the mechanisms outlined in Section V, Volume I of this report.

$$z = -\frac{1}{R} \left( \frac{1}{T} - \frac{1}{T_R} \right)$$
 (A-24)

$$G_1 = \frac{1}{k_{12}} \exp(E_{21}Z)$$
 (A-25)

$$G_2 = k_{2W} \exp (E_{2W}Z) \qquad (A-26)$$

$$G_{3g} = k_{3g} \exp (E_{3W}Z) \qquad (A-27)$$

$$G_4 = k_{4W} \exp (E_{4W}Z) \qquad (A-28)$$

$$G_{48} = k_{48} \exp (E_{48}Z)$$
 (A-29)

$$G_{\delta \delta} = k_{\delta g} \exp (E_{\delta g} Z) \qquad (A-30)$$

$$G_6 = k_{66} \exp (E_{85}Z) \qquad (A-31)$$

$$G_7 = k_7 \exp(E_7 Z)$$
 (A-32)

$$G_0 = k_0 \exp(E_0 Z) \qquad (A-33)$$

$$G_0 = k_0 \exp(E_0 Z)$$
 (not used) (A-34)

$$G_{10} = k_{10} \exp (E_{10}Z)$$
 (A-35)

$$R_1 = D_{10} G_1 \qquad (A-36)$$

$$R_1 = D_{10} - R_2 \tag{A-37}$$

$$R_3 = (G_{38}\gamma)A_1 \tag{A-38}$$

$$R_4 = (G_{4B}\gamma)A_2 \qquad (A-39)$$

$$R_6 = G_6 \gamma A_3 \qquad (A-40)$$

$$R_0 = G_0 \gamma A \qquad (A-41)$$

$$R_{7A} = G_7 \frac{Vf_A}{Q_A} [NO_2^+] A_3 ; R_{7M} = G_7 \frac{Vf_A}{Q_A} [NO_2^+] A_4$$
(A-42)

$$R_7 = R_{7A} + R_{7M} \qquad (A-43)$$

$$R_{0A} = \frac{VG_0f_A}{Q_A} [NO_2^+] A_3 \qquad (A-44)$$

$$R_{0M} = \frac{VG_0f_A}{Q_A} [NO_3^+] A_4 \qquad (A-45)$$

$$R_{e} = R_{eA} + R_{eM} \qquad (A-46)$$

$$R_{0}A = G_{0}\gamma A_{0} \qquad (A-47)$$

$$R_{\bullet A} = G_{\bullet \gamma} A_{\bullet}$$
 (A-47)  
 $R_{\bullet M} = G_{\bullet \gamma} A_{\bullet}$  (not used) (A-48)  
 $R_{\bullet G} = R_{\bullet A} + R_{\bullet M}$ 

$$R_{\bullet G} = R_{\bullet A} + R_{\bullet M}$$
 (A-49)

$$R_{10A} = \frac{VG_{10}f_A}{Q_A} [NO_2^{\dagger}] A_3$$
 (A-50)

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$$R_{10M} = \frac{VG_{10}f_A}{Q_A} [NO_2^+] A_0$$
 (A-51)

$$R_{10} - R_{10A} + R_{10M}$$
 (A-52)

$$C_6 = 11R_7 + 14.5R_6 + 9R_{10} + 14.5R_{9G}$$
 (A-53)

$$R_0 = (F_D) (C_0) \tag{A-54}$$

$$C_{8P} = C_8 - R_8 = C_8 (1-F_D)$$
 (A-55)

$$\tau_{A} = \frac{Vf_{A\rho A}}{A_{T}} \tag{A-56}$$

$$A_1^0 = R_3 + D_1 + M_1 - A_1 = \tau_A \frac{dA_1}{dt}$$
 (A-57)

$$A_2^0 - R_4 + D_2 + M_3 - A_2 = \tau_A \frac{dA_3}{dt}$$
 (A-58)

$$A_{8}^{O}$$
 +  $R_{8}$  -  $R_{6}$  -  $R_{7}$  A -  $R_{6}$  A -  $R_{9}$  A +  $D_{8}$  +  $M_{8}$  -  $A_{8}$ 

$$= \tau_{A} \frac{DA_{b}}{dt}$$
 (A-59)

$$A_4^0 + R_4 - R_8 - R_{7M} - R_{8M} - R_{9M} + D^4 + M_4 - \Lambda_4$$

$$= \tau_{A} \frac{dA_{4}}{dt} \qquad (A-80)$$

4.

$$A_8^0 + R_8 - R_{10A} + D^0 + M_8 - A_8 = \tau_A \frac{dA_8}{dt}$$
 (A-81)

$$A_6^0 + R_6 - R_{10M} + D_6 + M_6 - A_6 = \tau_A \frac{dA_6}{dt}$$
 (A-62)

$$A_{\gamma}^{0} + R_{10} + D_{\gamma} + M_{\gamma} - A_{\gamma} = t_{A} \frac{dA_{\gamma}}{dt}$$
 (A-63)

$$A_0^0 + 3R_{10} + D_0 + M_0 - A_0 = \tau_A \frac{dA_0}{dt}$$
 (A-64)

$$\Lambda_0^{\circ}$$
 +  $P_0^{\circ}$  -  $P_0$  -  $D_{10}$  -  $R_0$  -  $R$ 

$$-A_0 = t_A \frac{dA_0}{dt}$$
 (A-65)

$$A_{10} = 0 \tag{A-66}$$

$$A_{11}^{O} + A_{14}^{O} - C_{1P} - A_{8O_{4}} = t_{A} \frac{dA_{8C_{4}}}{dt}$$
 (A-87)

$$r_{114} = r_{01}p - A_{14} = r_{A} \frac{dA_{12}}{dt}$$
 (A-68)

$$A_{18}^{O} = A_{14}^{O} + R_{1} + R_{8} + R_{8} + R_{4} + R_{8} + R_{4} + 15R_{7}$$

$$+ 16.5R_{8} + 12R_{18} + 16.5R_{8O} - (.33R_{p} + .67) R_{8}$$

$$-A_{L} = \tau_{A} \frac{dA_{L}}{dt}$$
 (A-69)

If 
$$A_L < 0$$

$$A_{13} = 0$$

If 
$$A_L > 0$$

(A-71)

$$A_{11} = A_{80_4} - A_{14}$$

$$\tau_{\mathbf{p}} = \frac{V (1-f_{\mathbf{A}})}{P_{\mathbf{T}}} \rho_{\mathbf{p}}$$

$$P_1^0 + R_1 - M_1 - D_1 - P_1 = \frac{dP_1}{dt}$$

$$P_8^0 + R_8 - M_8 - D_8 - P_1 = \tau_P \frac{dP_8}{dt}$$

(A-75)

$$P_8^0 - M_8 - D_1 - P_3 = t_P \frac{dP_8}{dt}$$

(A-78)

$$P_4^0 - M_4 - D_4 - P_4 = \tau_p \frac{dP_4}{dt}$$

(A-77)

$$P_0 - M_0 - D_0 - P_0 = \tau_P \frac{dP_0}{dt}$$

(A-78)

$$P_1^0 - M_0 - D_0 - P_0 = \tau_P \frac{dP_0}{dt}$$

(A-79)

$$P_{\uparrow}^{O}-M_{\uparrow}-D_{\uparrow}-P_{\uparrow}=\tau_{\mathbf{p}}\frac{dP_{\uparrow}}{dt}$$

(A-80)

$$P_0^0 - M_0 - D_0 - P_0 = \tau_p \frac{dP_0}{dt}$$

(£8-A)

$$P_{10}^{0} - D_{10} - P_{10} = \tau_{p} \frac{dP_{10}}{dt}$$

(A-82)

$$P_0 = 1.51 \left( \frac{A_{11} A_0}{A_{11}} \right) \left( \frac{P_T}{A_T} \right)$$

(A-83)

$$G_{CO_{1.8}} = R_{10} + 6R_{1} + 7R_{0} + 7R_{0}G$$

(A-84)

$$G_{NO} = .67R_0 (1 - R_p)$$

(A-85)

(A-86)

# Appendix B

Dynamic Simulation--Program Listing and Output

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PROGRAM THISIM 74/74 OPT=1

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	PROGRAM TRISIN	74/74 OPT=1 1=190 A/A	11/15/74	13.13.41.
<b>\$</b>	CALL 9	CALL SUP If J.WE.NH. 6070691 WPITE(65.509) FORMAT( 12M9SEPARATOR 4)	1810v 1810v 1810v	;;;;
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Ş	P45(1) #P 41 1(1) #A5(1) P P (1) #B5(1) P P 1 (1) #F (0) P	)     -  Facion=14<     -  Fre=145     -  Fre=15=155    -  Fre=15=155    -  Fre=15=155    -  Fre=15=15=15    -  Fre=15=15=15    -  Fre=15=15    -  Fre=15=15	TMTOV TWTOV	
Ž.	700 CONTINUE 1F (101)1T CAL. SUB 1F (14,005)		VOTAT VOTAT VOTAT VOTAT	5555 5
\$ 8	510 FORMATI 11 WRITE (6.40 CONT WEE FOREST (6.40 EAR-EARY WRITE-WES FOREST (6.40	FGCHATE 11HQMITRATOR 5) MRITE(6.401) CALL STRICK.POAI.PI) EQD=EQAP5 EAPEAQP5 EVERFEVENS	TMT07 TMTD7 TMTD7 TMTD7 TMTD7 TMTD7	3848685 3848585
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	p(1)=p6(1)	THTOY	F.
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	720 CONTINUE		: X
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	CIS CAMPAT INVESTIBATED AS	THTOY	2.
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	_	THTOY	<b>1</b>
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	F0966=1F06	THTOY	261
25	FACTP6=1 - FA6	THTOY	285
	1F(1, MF. 144) 6070731	THTOV	563
	WPITE (6.517)	THE	Y.
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e i e		THTDY	201
Ê	CANAL CHANACO CHOMACO PERMACO OF PRICO	TWTOY	286
	247 FORMAT (TRINGCONVERGENCE TESTS./ABA.4HACTO.4X.BHORGANIC./	TNTDV	284
	_	TATOT	210
	2 6X.46444XIMM PEPCEHT CHANGE FROM PPEVIOUS ITERATION-3X.2F10-33	TATOV	211
516	WITE (6,246) (1,1=1,6), FACID-FONG-FRC	TATO	512
	246 FORMATE ABH-DEARMETERS USED IN RECYCLE CALCULATIONS/AL SYSTMME.		517
	C 271616/61 34/FBACTION ACID PHASE IN ORS. SIME MICH B.S.	THETON	515
	TO STREET THE LINE FOR THE PARTY OF THE PART		•

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	J	61 254FRACTION INTERNAL RECYCLE. PAGE 0.51	1910	
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			A. C. L.	•
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CONTINUE  WITE (6.46)  WOTTE (6.46)  WOTTE (6.42)  WOTTE (6.42)  WOTTE (6.42)  WOTTE (6.42)  FORMAT (1) **ALM (1) **	7		TATEN	
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C		200 and 14.50 3/ 04 000	TWTDY	
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FORMATE 19-6-5X 21MEXTESNAL ACID RECYCLE)  WPITE [6-40]  W	•		TNTDY	•
### ### ##############################		•	THE	CKS
WPITE(6.40!) WPITE				•
WPITE 16.467   ARR. (PRR([].1=].10).AIM(2).AIM(2).AIM(2).AIW(2).   THIDY		WPITE (6,401)	10141	,,
C ATMIS, THIES, AND LESS AND L		NOTTE (4.442) AXB.(PXR(1).[2].]6)6.ATM(2).ATM(5).TM(2).4TW(2).	TATOY	ur.
INTEGE   146.5X   21   21   22   22   23   24   24   24   25   24   24   24   24	•	TO ME TO THE CONTRACT OF THE C	TNTDY	
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FORMATIC 1146-5X 2 IMINTERNAL ACID NECYCLE)  WOTTE (6.462) A [A. [A. [A. [A. [A. [A. [A. [A. [A. [		<b>VRITE(5.464)</b>		•
### ### ##############################	ţ	FORMAT( 140.5% ZIMINTFRNA, ACID RECYCLE)	IMTOT	,
UPITE(6-402) A[R-(P[R(1)-1=1-10)-ATH(1)-ATH(1)-ATU(1)-ATU(1)-ATU(4) THIDY CONTINCE CONTINUE C	1	112 (4.44)	YOU W	
C - 1 L (1) - A		・・・・・・・・・・・・・・・・・・・・・・・・・・・・・・・・・・・・	TRYDY	•
CONTINUE C. *IMID: AM_(1) -AM_(4) -TW_(1) CONTINUE D2748[=1,4] A65(1) =A(1) A65(1) A65(1) A65(1) A65(1) A1TOY CONTINUE CONTINUE CONTINUE CONTINUE CONTINUE CONTINUE CONTINUE THEO THEO THEO THEO THEO THEO THEO THE		TARTES TO SELECT THE PROPERTY OF THE PROPERTY	ALT.	
D3746 = .14	_			,
D2748 = .14	216	CONTINUE	TATOY	,
COTANT   C			TATOY	•
A65(11=0(1) PVT(0) PVT(1) F((1,4CK/MP)**PME_JACK)60T03 F((1,4CK/MP)**PMP_ME_JACK)60T03 F((1,4CK/MP)**PMP_ME_JACK)60T03 F((1,4CK/MP)**PMP_ME_JACK)60T03 F((1,4CK/MP)**PMP_ME_JACK)60T03 F(MTDY F(MT)=(F065(3)*P65(4))*F06**PMF_ME_JACK)60T03 FMTDY CONT ((COUNT)=100**PMF_ME_JACK)60T03 FMTDY CONT ((COUNT)=100**PMF_ME_JACK)60T03 FMTDY FM		11.1=10.103		•
PATOY   PATO   PATOY		W65(1)=A(1)		,
	746	P&C (1) ±P(1)	TWIDY	•
CONVECTOR   THE CONTROL   TH	?	Coordinate the Control of the Contro	TATOY	25.0
			VILLE	
f0x7=(f0x5(3)+0x5(4))*F06+(4x5(3)+4x5(4))*F4C[D6)*182* [WIDY SUM=0. T0 75 1=1.9 SUM=SUM-(4x5(1)*FAC[D6+0x5(1)*F06)**** [WIDY CDx1(1COUNT)=100.***********************************		[CON•1 = [COUNT • ]		
\$\text{SUM=0.}\$\$\text{SUM=0.}\$\$\$\text{SUM=0.00}\$\$SU		1047 = (1265 (3) +P65 (4) ) +F06 + (265 (3) +A65 (4) ) +FACID6) + 162 -	14101	•
TWIDY SUBJECT   1   1   1   1   1   1   1   1   1			TATOV	
CDG 75 1=1+8 SUR=SUR-(A6S(1)+FACID6+P6S(1)+FO6)+MP(1) TWIDT CDG/1(ICOURT)=100.+TDMT/SUM TW 1=0.14 TW 107 TW			THIRDY	•
SINESQUE (ASS(1) + FACID6+P6S(1) + FO6) + PM (1) THIDT (COURT) = 100 + + + + + + + + + + + + + + + + + +		-		•
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AFRONU(1)	THEO	578
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AFRONALT)	TATOV	3
	TMTDY	3
SPENT (1COURT) = SPA	TMTDY	5,42
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P+62,4)	TMTDY	ŝ
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- 35-36-37-385-1	THIOT	
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orlass.es.	10111	5
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ef0861 ef181 e01, T	TOTAL	2
	TOTAL	Ş
DG07=110  DG07=110  DG07=110  DG19=120=55(J-1)7800(J)  DG14(1C-1)=PP/(QP+62-4)  DG14(1C-1)=PP/(QP+62-4)  DG15=1-R  AM4(20-4)  AM4(20	ETV-F BN(S-110-EQL10-252	·f Ints. 1) •Eq. 1 •.252

-	PROGRAH THTSIN	PATSIN	74/74 OPT=1	FTN 4.1+PSC367 1	11/15/74	13,13,41.
2		E E	0=0-ENTH(A)S-FA2(D)+1-,PIS-FO1-11SEP(1)) +ENTH(A3S-FA3-FR3-P3S-FORG3-FR3-FSEP(3)) +ENTH(A2S-FRAZ-FRAC2-P2S-FORG2-FRAC2-1SEP(2)) +ENTH(A2S-FRAC2-P2S-1172)		40121 40121 40121	5586
ž		Ų U	GOTOSO GOGENTHARSS.FACIOZOI.,PPS.FOZOI.OTSEP(2)) HENTHARSS.FAGOTOSOI.OTSEP(A)) -ENTHARSAI.OTSPAOI.OTSAI GOTOSO		TATON TATON TATON	15353
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š		ი <b>°°0</b> ზ	CWIK(46+1.*1.*P4.1.*14) GOTUS-B  0=0.EWIWIA4S.FACID4.1.*P4S.FO4.1.*TSEF(4))CWIK(46S-FA6,FP6.FO7.FORG-FP6.TSEP(6))CWIK(46S-FA6,FP6.FO7.FORG-FP6.TSEP(6))CWIK(45S-FA6,FP6.FSFFA6S-FSFFAGS-FFAGS-FSFAGS-FSFAGS		TMTDY TMTDY TMTDY TMTDY	<b>13 13 1</b> 3 5
\$		* * * * * * * * * * * * * * * * * * *	601050 0=0.ENTH(ASS.FACIOS.i.,PSS.FBS.i.,TSEP(S)) -ENTH(ABS.FAS.FRACO.PSS.t.ORGO.FRACO.TSEP(6)) -ENTH(ABS.FAS.FRACO.PSS.t.ORGO.FRACO.TSEP(6)) -ENTH(ABS.I.S.FRAS.I.S.F.S.T.ORGO.FRACO.TSEP(6))		TWTDY TWTDY TWTDY	35253
\$2			00 678 [m] 66 66 679 [m] 66 679 [m] 66 679 [m]		TATOV TATOV TATOV	7.555 7.555
3		678	qa=na+a5(1,1)/RHQ(1) Pa=pa+a5(1,1)+RH(1) Polyski(1c+1)=pa/(04+62-4) CONTHUE WEITE(4,6) (COMI(1)+CO+DE(1;+COMSa(1)+CUSA(1)+COMTSa(1)+TCOM(f)+	DWTSA(1) .TTCOK(f).	TWTDY TWTDY TWTDY TWTDY	3 3 4 8 3 3 4 8 9 8
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8		~	C .J=1.IC) FOCATICLE.ZAK.eACTUA MITRICE.SEN.eSPECIFIC GRAVITYE/INIG.SILE. FOCATIOLISTAR.eAcTUA MITRICE.SEN.eSPECIFIC GRAVITYE/INIG.SILE. FOCATIOLIST (I.I=1.6.).A.A.A.G.H.S.L.A.B.D.B.L.B.L.B.L.B.L.B.L.B.L.B.L.B.L.B.L	RAVITY®/1X16.5110. ) RECYCLE FROM EACH	TMTDV TMTDV TMTDV TMTDV TMTDV	75 55 55 55 55 55 55 55 55 55 55 55 55 5
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	_	<b>*</b>		ENOTH	2
175	CH NOSY-I	ij		TMTDV3	æ
	CH256425-4	4-5-1		TWTDY3	Z
	X-3=100013	F-X		TWTOV3	2
	CHEWEN	CH20=H+X-{Y-X)		TMTDV3	<b>7</b>
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Ī	¥/1=0	•		141043	<b>B</b>
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	·	IF (189.6750414) GO TO 2337		TWTOVA	
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	CH2504=8+F			THYDY	-
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Ē	CH30=0.	•		TMTDY4	<b></b>
	AET=CMO2=GA	102=0A		TNTDY	2
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		12.		THTDVA	7
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2	C SULFUR	SULFUPIC + ALEUM SYSTEM (REGION 3)		TNTOVA	91
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SUPPOUTINE SUB	8	74/74 OPT=1 FTR 4,1+PSR367	11/15/14	13,13,5
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		AIT=CMC200A	THIBY	2
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	U	COMPUTE DEACTION RATE MEFFICIENTS	THIBY	Ĭ
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	_	CMSTTE(4.381) DELTA-FVKM,FA.ZD-SA-CB-A110A-SC-0A-63(IV)-535(IV).	THTDY	1693
245	_	GABER P. F. T.	TWTDV	1
	Ħ	FORMATION DELTA-FUKH-FA.7A-SA-CB-Allon-ElS.6. 6F10.4/	TNTDV	2
	_	C= 5C-04-63-635-64444-670*-2710.4-2815-6-710.6-715-4	THTOY	2
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		4	THTOY	101
Ē		)T-2427		2
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SUBMOUTINE SUB	8 74/74 OPT=1 FTM 4-1-498367	11/15/74	13-13-53
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U		TATOT	123
		TATOV	2
	MACKWARD DIFFERENCE INFERNATION OF ORGANIC SPECIES	TOTAL	201
U		TATOL	1631
	DTP-OP-01/(FWXHOFP)	TATO	7535
	OTA=OA=OT (FWM=FA)	TATOV	1633
£	PIO= (PIO-DTP-PIIO)/(IOTP-ETAIL-OTP/PME)	TATOY	*=
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,	A2=(A12 -C1012) (F4-C2)	ACLMI	2
	3	TOTAL	1621
	Past 1012	TATOV	25
	725.674.694	TATOT	1653
315	2=7/12-(7)	TOTAL	Ĭ
	12A11-P13-R3-2 - (A13-R3)	TATOV	55
	YEZ-CI+1	TATOT	256
	TJs(T)-07Pex)/(1,-07Pex)	TATOT	1521
	A3=(A13+03+(X+T3)/(F5+F7A+F8A+F94+C2)	TOTAL	25
320	IF (A1,1.1.8.) A3=0.	TMTOV	2
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r A		VIOLERS OF 164	•					
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	_	r=2 •CI+1.						
	-	TERITORING TOWN	T1011/(I:	Ī			TATOV	Ĭ
2	•	16= (A14-86+C3-76) / (4.or 108-C2)	CX-763/16.	(Z)-#617-			TMTDY	Ĭ
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	0		1416-PF-06-	[8]6.84.06.844-8] <b>9</b> 8-46]	_		Tator	13.16
Ž	•	047= 1	(A17-A#T-#10-A7)	10-17			THTDY	119
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	•	367	-67-60-1de	12.500063			THTDY	1127
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		COMMON (**)	CHRONITIME SURL(A).PO.A].PI) DIVERSION BEALLA:DEP(14) DIVERSION AD(16).PD(16).PI(10).AI(14) COMMON [X.D].IPRNI.JACK.NM.MM.TIME.ICALL.NP.TCOX;INOX.RATES(8:10)	P P P P P	VCT V VCT V VT V V	1233 1234 1236 1236
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Ì		36. PM. 36.646 8174.874		MUM	, 66 7.	He SOO	1679.783	11.45			900	ORC. PH.	5.676 1266 206	13.988			HDAT	0 6	9	7.	A445.131		1204	000	100.	900		16.055		Trical		980	0.	654.311 7.191			In Com	6	0.000	22.6	5137.466	•
STAGE	2 5 5 5 5 5 5 5 5 5 5 5 5 5 5 5 5 5 5 5			ADM	114.	•	<u> </u>			AQ4.	684		R A	. Z		T STAFE			- 1	74.3	<b>£</b> :	2	Track	100	-	ż	2	15		1000	C 10.		5	3 t			A(P'aT	90	į	.104	ř.	g E
TO MENT		4510 Pt	e,	DE CYCLE		ACID PY	150.00	141.11	PECYCLE			ACID PH.	186.5	165.585		I TO MEET		500	100e	1	69.198	ę	PECTAL	6	900	ACID P	150	14945.	Œ.	7	0 0	AC10 PH	47.55A	6291,246	•	10 WEAT		3	AC10 PH.	•	9.075	•
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SEPABATOO 4 ORSANIC	į		10. CM	FATERN	f £	!	200,65	A CHINE	TATEMEN		#C (1) PH.	•	ES FC	VOLUME VOLUME	2 001101				ř.	MCL FS	5580	A.S.	F £ 75 3N	d		,	21.FS	AUTUAE SUTUAE	1. " f		THE C		POLFS	VOR UPPE	A DOTAGO	06.50.10		ECTO PH.	į	Salibe	MAS.	A
SEPA	AC10	į			AC 15	,					1	,			2000	<u>.</u>		9119	9						960						AC 15				5	,		172				

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	141	562*	.367	TOTAL	166.482	15757.726	156.137		MINI	650	£5.	TOTAL	27.73	2726.955	27.366
	ATNT	4.R20	7.224		~	•	_		ATRE	1.189	1.25		•	~	•
	#Ox1	. 690	000.	086. PM	7.56	1715.36	18.711		1 Patch	1 000	.000	0865. PH	1.36	296.85	3.23
	ADMT	.003	26.0.	:	2	'n	£		AUNT	. 001	200.	÷		21	2
PECYCLE	Terre	000	.900	ACID P	152.R	14642.34	133.4	PECTOLE		.000	.000	ACIO P	76.45	2430.11	24.128
AL ACID	PERT	200	000					AL ACID	Tr.WW	200.	.069				
EXTECNAL ACI		ACIO PM.	COG. PH.		FOLES	SSVH	ACT DA	INTERNAL ACT		Acin De.	TOC. DH.		#M.ES	MASS	Jack TOA

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15162.02	25.101.27	15102.30	15103.05	15103.03	3514.35	15115.	15105.77	15105.81	15165.64	15105,43	15105,07	15104.66	15164.21	36.791	15161.77	15105-15	15104.75	15116.0.	15128.71	15148.45	15176.61	5213.55	15.00.51	15377.65	15444.87	15414.99	2.585°	15655-17	15761.61	EL 01 131	14,000,35	15433.36	15971.33	16603.58	16652.95	16071.19	166#5.89	16047.65	70.00	16119.45	14123.56	16175.59	14174.75	161 36.22	16131.16	16131	16131.06	16131.54	16131.28	1~1 30. AZ	14130.31	161 29.78	55.55.141 55.151
70.11 2.10.1			1.4732	1.4732	1,8733	****	46.4	1.877	1. A7.X	1.874	1.574.1	1.174	1.676	92.4		100	9,44	1,146	1.4329	1.A.75	1.4028	1.9647	629	10.0	256.1	1.9466	1.9562	<b>X</b> • • • • • • • • • • • • • • • • • • •	9870	1 9866	ī	2.0016	2.5642	7.0168	2.0254	2.0123	2.0375	2.5423	7.00	2.056.1	2.0591	2.0427	2.0631	2.9693	2.675	2.0781		-		-	2.879	2.07A1	2.0776
11001 4.034)		10.0	A.e.33	4.e396	B.8399	70-0-0	9199	4.6415	0.6.10	2.1.1	15:9.8	F.0.8	2			B. CA16	B. 69.36	9-14-0	8.1235	4.1.3]	4-1459	81:1:0	1417.	A. 2418	9-3156	8.3504	8.3457	R.4212		5.26.3	9755	5475	9.6170		4,67.4	8.7298	4.7-21	2 2 3		B.BIRI	R. R. 76.6	8.8591	B.Ank7	8.8786		6.0142	A. 92 30	₩.O.	A.4753	A.4733	4.9701	A. 9168	5.9112
10.77		13.73	14.726	19.712	19.49	47.0	10.00	19,653	1.5	16.437	16,631	10.675	14.461	19.61	-14-41	16.612	19.414	19,419	14.433	14.44	14.476	19.786	- 1 - N - 1 - 1	- N	19.934	20.006	20.0%	<b>3.15</b>	25.23	241.04	20.00	20.514	20.578	26.64	652.02	20.417	20.07	20.933		31.11	21.171	•	-	21.359	ᆣ.	~ -	21.412	: -:		21.834			21,043
22.55 22.95 30.95		25.72	22,964	160"LZ	27,014		21.846	7		27,647	53.0	21.077	23.041	53.645	21.081	21.688	21.045	21.075	21.654	21,629	22.487	22.431		27 673	655,55	22.439	27,315	22.199	27.051	710 16	21.733	21.639	21.553	21.477	71.17	21.304	21.242	21.227		35.	21-15	21.13	21.12	21.17	21.113	21.11	711.17	71.17	717117	21.171	21.13	21,131	21,137
.1984		13051	15051	6764	14661.	466	11001	19015	21961	16001	19329	62061*	*2051*	1605	47CA1	200	19942	94.64	I CO .	10116	5111.	1633	K-56-51 .	10401	19763	19933	*1102*	5020	4707	69702	21019	67117.	.21323	21466	70117	21412	.21 +67	-2166	61827	81000	11444	173.76	A7155.	52762	27478	21527	275.88	12.27	45467	22.48	H1147	127767	24175.
5269.8	2000	5200 a a	F 462's	5.99 s. B	8 4025	2.00.2	1706	1 V 0 V V	5155.3	5176.1	5042.2	5033.1	4.757.5	5.4169		48084	1. 1484	4.7.4.	4.54.5	4,642.1	4.64.4	4.000	2.6.44	46.75	17.4	1.01.0	4424.1	6.25	4832.7	1 4 3 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4	4453.2	4.14.3.5	4.675.3	0 m/c.	10.00 P	26659	44.3.	5.450.	2000	C	6.576	5.914.7	1-2202	5632.3	30°00°0	2.8375	410707	5 6145	6.7.92		2-5-5-5	9.455.5	
79.396 29.396		STERM.	25837	-20405	.75413	17707	20.31	70466	20502	.23411	50-12	.22103	44622	-23175		20013	25.55	4	25-45	21.2-7.1	27.543	35-56	17.7. A	37731	- CC-14	24334	.22567	\$ [ • • C	\$5.55 \$7.55 \$7.55	74.47	0126	F750E.	.35-34	9-116	71.17	1441.	1822	.3745.	. 32-50	A 4 5 4 4	1 d 1 d 1 d 1 d 1 d 1 d 1 d 1 d 1 d 1 d	174.34	211.	33: 4	1.44 L.	3.53.6		1619	9		- 11 -11	7 7 7	4 4 4

14121.17	16124.62	16127.72	16127.48	16127.27	16127.10	16126.97	16176.00	16126.73	16126.76	16126.75	16176.78	15126.42	16125.88	16126.95	16127.01	16127.05	16127.06	16127.04	16126.98	16126.97	16176.71	16124.56	14.126.25	16125.45	16175.61	1612.2	16124.84	14126.41	16127.96	14123.44	16173.43	16122.53	16122-05	14121.56	16121.09	1979791	16120.15	16119.70	16119.23	16118.63
2-6745	2.072	2.0719	2.0707	2.86%	2.638	2.0A77	2.8564	2.1463	2.056	2.0455	2.6453	2590.2	2.8452	2.0-54	2,0%57	2,0640	2.3445	2.0%79	2.8675	1890.2	2.05AR	2.8694	2.1701	2.676A	2.1715	2.0.2	2.0730	2.0737	2.0744	2.0751	2.675	7.0.Z	2.071	2.8778	2.6784	2,174	2.17%	2.1482 ·	2.1617	2.6413
7.004	1540	£.0597	0.4545	B. 8797	R.9753	A.4715	B. PAR?	A. 9655	* F	9.8619	B. Ph. 11	8 . n6.86	6.14.0	1176.8	24.0	7. ** · E	4.44.3	1. 46.AL	0.0764	1.17	B. A7¢ v	A.479.	4.1819	4.4844	9749.2	A. F018	1.65.1	1460-6	¥.0862	6,4432	F-98-3	1.00°.	F.911B	1.914	6.4173		8.9275	B. 9256	4.4774	16/4.8
22.184	22.150	22.787	22.22	22.24	22,331	22.36	27.34	925. 55	22.441	27.450	22.473	22,483	22,699	22.45	22.497	22.6%	27.44	27.49	22.445	22.4AE	22.474	895.55	22.462	77.451	2552	27.648	577.22	22.443	22.441	22.41	27.42	27.443	22.44	22.450	25.454	27.459	22.405	22.472	22,479	27.487
21,151	21.163	21,176	21.186	21.14	21,213	21.224	27.24	21.260	21.276	21,293	21,311	21,324	21.3%	21,343	21. 185	21,347	21.413	21.42	21.444	21.459	F117	21.464	21.499	21.589	21.52	21.534	21.533	7. Y.	21.554	21.763	21.579	21.576	21.542	744.15	294.15	21.597	71.641	21,685	21.M84	21.612
22824	22849	15455	22892	.2241	£2624°	77522.	65622	.27971	. 729A3	₹ <b>?</b> ?	.23033	-23012	05CF5.	.2 1077	FE6E5.	£262°	77312"	64367	\$20.2°	-23059	.23963	8-CL2-	67065.	71015-	-23042	MF-01.2*	.>3043	PC 96.2°	-23184	11116	11167	£51r7.	2117.	AF 11 5.	25162*	67162	23155°	53165.	.23144	.73175
-135	5654.2	5054.9	5055.0	5957.3	5059.	5351.8	5643.3	5005.8	506H.6	5671.5	5.077	5.07.0	5641.3	5.0% Z	50FE.1	9.10.65	5635.1	5659.5	S161.9	5105.3	S184.5	5111.7	5114.8	5117.7	5129.5	5123.2	2.2.	5126.1	5136.4	5132.5	514.5	7. S. IS	5134.1	9. Ff 1.4	5141.3	5142.7	5144.6	5145.2	5144.4	51.7.5
47.176	*	12.3	3-671	34044	34020	11400	.33441	33300	31352	337.5	33512	33124	Plett.	3,413	33310	TUALL.	30116	,33703	33401	13600	P. 9.34.34	13465		OFTER.	. 33544	.3341	33475	33443	1 3756.	114.1	37444	Eirle	64.56	.1+1£	33767	33754	-3375G	23754	.33747	33175

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7-171111	111157.7	111207.5	11225.1	111275.4	2.765111	111347.0	100054.6	13/24/00-2	114417.1	102440.2	6-17-4 6-17-6-4	1.06170	A2797.7	A1763.0	6.647	A0116.5	60267.1	A654.2.3	#1851.0	24.45	#357R.3	2.14.74	1.501.19	8419.5	4.50.16	93044.1	74.375.3 66.74	4.663.7	47461.3	X	100570.6	101465.4	102855.5	103471.8	18442.8	10505.7	105532.1	105007.1	1641/6-7	118617_3	1153117.1	11#27.5	120465.2		7.14.71
7 121196.0	27175	321555.1	121646.0	321405.6	371465.6	1770	174346.3	7.0101.7 7.57.7.7	174855.A	375451.0	374600.0	364112-3	342 341 .9	359167.9	155427.B	1546	7.000	75.076.7	156371-1	× 5 34 4	4.767.4	X2417-17	172753.	375524.1	141475.0	345075.3	JARBAL.1	703660.3	746756.7	4.540147	403764.6	4.05320.0	0.02100	4.1 Bab 7.t	412512.6	415535-2	416926.7	2-132614	H 4 4 1 1 1	21163.9	419h72.4	414447.9	4. 7. es. e.	2"280417	0.010014
4.9116.4	1.17479	977628.6	977124.0	97743.4	4776-1.4	*******	4. +10170	4 Ct CS	B. 6.70.40	1666241.A	1384843.E	1011 100.2	1011479.0	1010743.7	1 004427.A	1004471-6	1.000	1616324.0	1013442.4	1019365.6	1023297.2	1027547.7	1034631.6	1041257.4	1958 761.1	1656731.0	1053045.0	1067627.6	10707-7.9	- CL87741	1041124.	1004721.2	1000015.2	1692715.7	1005746.3	1100102.9	1102155.4	1104514.0	1105546.4	1117490.4	1115244.5	1114765.1	1117570.2	1114077-1	1119-4-7
5 886842.3	Pulled.1	A61264.3	AF1231.6	PA1276.A	PH12.4.3	A-1330.4	Nal 345.7	2-541 1-4	na jaca, y	AH2779.4	445285	7.64	F-01 75.2	4-141-4	d out tolk	# 17.00 M	A-2331.4	P-14-14	200	B-17176-B	A-502.7	F. 7.25.	R-2413.0	8-1174A.0	1 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4	1.981078	F78596.4	47.74	A7nd72.9	87665.8	A7577A.1	675427.5	67555	874717.A	874535.0	P742-2-3	A74127.1	674631.3	677452.7	6.15.74 6.74 14.74	1.16.574	B17576.5	P. I Hotel	F7   746.B	R. 1. 1. 1. 1
2.1570851	R-5949651	9°9119821	176965A.3	178590	D'Alangel	126-417.6	120464.	1/07/17:1	1211-94.4	1215119.5	12196-0-6	120145	1227077.0	122-143-9	1773440.1	121542121	171070171	3.785. 784.A	3.00145.0	1176415.5	1144463.0	1131975.0	1174=32.4	1171754.5	110447011	1194111.4	1161001.4	- 52.25.11	1150564.6	1157-611	1157111	1150414.3	1142419-8	1147315.9	11-6275.6	6-1-4	1143444.3	1142462.7	11-14-5-7		11 3-6-0.2	113777	1134 2.00-1	4,421-11	9°21 36 11
3 2068092.4 2661616.1	2051675.3	2462664.0	2062979.2	6"224.482	2045454	7794676.9	2644452	204	269-114.2	5-1-5-96	26-75-7-1	207.707	237-112-2	20457-7-B	E-7.1957	2112434.6	2122.75.9	1. 1411115	2134372.6	2150000	215- 120-1	2162401.3	214455.7	2171-466.0	2175364.2	21765413.5	71.75%.9	1. EE way 1.2	2174247.3	2174737-1	71 74-04.R	2174-04.5	2179574-8	2170355.5	2174975.3	2.946.15	2117413.2	5177435.2	2170726.0	5, 14, 2, 17	21/01.7	2117715	21 71 300.4	214	4.00 JAIL
2 \$89895.5 \$48618.1	544264.5	544334.2	584582.5	**********	2"11165	# <del>*</del> # # # # # # # # # # # # # # # # # # #	9*Ft yr #5	7.47.54.17	9.17.6095	4-1411.6	598611.2	A. [ 44.4 - 4	H. 401 1.4.7	5.4534.5	Z*6515+3	A 100 A 100 A	5447-33-5	1.00-5rac	0.6-5-5-5	10000	9.4. A. D. S	597]P6.8	59-642.7	504175.6	5.44.3cm	4.4.4.6.9	9*418.07	601126.7	£61477.3	6.00 Per 0.00	602749.2	5-12-19	66 161 3.4	6.624233	687463.1	667595.3	502421.7	\$62751.2	AC2C75.2	7-00-104	1.655103	A	P.1411CA	40.500.00	6. 6. 6. 6. 6. 6. 6. 6. 6. 6. 6. 6. 6. 6
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125236.4	12751.2		176722.2	126432.2	127164.2	17754.4	127344.0	127534.4	1274-64.5	127764.0	127793.0	127477.1	127057.2	124033.7	124107.0	12a176.9	124243.1	124705.4	124763.5	128417.2	1.45461	128510.3	12+5+9-6	124586.2	128-14-0	12**33.4	122440.5	122477.5	17-46-6	12-766.1	122764.2	124789.1	128769.8	124766.9	12-106.2	124491 .R	1280.80.8	120467.4	128651.6	124633.5
4.64701.	4.00404	1,57174	C. 28%	423180.2	423737.6	676776.3	47443.E	47113.6	**************************************	4.75470.7	4717.V	6.3.5.e. 7	475662.3	6,71165.5	4.72.57.4	4,27774.5	2.800462	4.045464	4/15/19.2	2.827.EC4	1.TAOR27	4,79205.1	5.51.45.	479403.2	4.267624	429471.2	4-10135.6	6.18/0F4	6-16-37-3	430572.8	6 36504.7	1.7[201.7	438922.1	431920.0	9. FEI 11. 2	P.P. 11 1F4	431250.2	431323.6	431377.4	431423.5
1110507.1	1120182.2	1128477.5	1124453.0	112377.2	1125413.2	1126311.7	1124445.8	1127514.3	112-14-22	1162440.2	11 Parsage	1130366-1	1131143.3	1131913.2	1137645.1	1133454.8	113-11-2	11 36475.0	1135727.1	1134656.5	1137174.2	1137479.0	1134554.1	1134746.7	E° /6*611	11-653-7	11-1155.5	1141750.6	114747.0	1142411.2	11+3448.7	1143647.3	31-45-0-	11-5n02.0	4.784241	11.5441.3	1144.794.8	114411	11-7220.5	1147413.0
A78792.6	N76427.3	1,000,00	669399.7	869135.6	854927.7	1-9H/848	364647.4	8-9c46.R	HAA537.7	P44739.6	46,00 37.0	164964.	869113.4	A69256.1	PAS-54-3	Andreh . 2	Anga37.5	M70031.3	A78226.0	478419-3	A70610.6	A787~9.8	178445.1	271164.1	A71344.2	A71575.3	A71. 1. 1. 4.	P11472.2	R126-2-3	R12210.8	872317.5	472541.3	A.17171A	R17076.7	T_FFOFTE	873135.6	#73357.3	A73514.0	873674.5	673438.5
1130 300.E	112479.4	1127193.8	115757.3	1125311.5	1124478.3	1124491.4	1124442.3	1124274.1	1126152.1	1124015.4	1123-221.6	1123064.4	1123740.	1123774.5	1123-75.5	1125-12	1123541.	112754.5	1123519.6	112 1:A7.6	1123457.3	1:27470.3	1123465.4	1121146.0	1121159.7	1123754.3	112 (751.3	1123764.	1123752.4	112142.5	4.777511	1123-64.0	1123524.1	112 2462.6	2"1271	11/3550.0	1122~54.5	11234.4.6	1123770.5	1123*03.2
2164419.3	2162636.3	2160030.7	2157203.1	2155-21.1	2153092.3	215-2033.9	2156445.4	21-404-13	2141775.0	2100 307.3	2145731.1	214-646.A	2143444.8	2143269.3	21-2776.4	21-7345.0	2142100.1	7141 403.4	2141/42.5	21-1725.5	2141720.4	2141756.4	7101923.4	2141712.4	9.216.415	2142176.1	# at // # 1 /	21-2767.4	21.2.49.4	1-14/-17	21-24-72-0	21-4-42-12	2142774.2	214770.5	F. Barl 5-15	21-2701.3	21-277H.L	214<750.5	21-2764.5	2142553.6
2.569869	669580.7	505474.0	600517-8	400250	668210.4	4.00189.5	466171.5	665171.5	e9:1+1.6	4.00221.5	A90273.0	6463-2-3	£30424.8	6.03530.9	600047.0	505774.6	501411.2	99164	A01290.8	6613-A.3	6.014.4.04	601674.5	661777.5	+01919.5	F.278 15.7	602145.3	49.7744.0	4677.50	6-65-43-9	40.746.734	1.65-584	4224.45	407 7 4 6 B	\$625534	SF = - 7 - 3	4.0 .6.200	632 1. 3	1.404.66+	69 10 31.5	533050°+
1893693.7	1443169.5	1803177.8	0.000	1842745,3	184244 1.6	1A-2121.9	14.17.1.5	1441241.1	Latition.	4.12F 2. Al	1 3- 4477.8	1245736.5	D. # # 7 F F F B	10-2-64.6	Part Creke	1417411	1A-17-7.2.	Mary 7/4 . M	1eer 5/1.55	143-	147511309	16-1714.7	IMMS7-1.1	Hand A. Andrew	Harry College	151	Irmy 179.7	1.5-2.4.1	14: 5]+i.B	1.50	1 Ent 7-4.1	1 7 37 2	2.11.	* a-1 . 3-0 !	2.4	*********	15975-2	157.1.3	19-111-91	14-6242.1

	*****	7	1.44937	1.44917	1.44937	1.64931	1.44937	1.44937	1.46937	16697	4694	1.496.10	1.44.935	1.46934	1.46933	1.46932	25047.1	156477	1.44930	1.44430	1.44929	1.46928	1.4497A	1-4-927	1-44927	120.00	1-46924	92077	XC 97 1	44974	1.44974	1.44423	1.44.23	1_46972	1.46922	1.44.921	1.64971	200000	46.047	01047	1-46919	1.46919	1.46914	1.4491	1.44918	1.46918	11695-1	1.46417		41044	1-44915	1.44916	1.4.916	1.4.916	1.46914	1.44914	1.44914
	2		4674	1.17	1.46784	707	***	1.44794	4574			.66783	3.647#2	1.1.1	1.477	1:46776	1.44774	1.47.1	16769	1.46706	1.13.1	1.46702	1.46704	1.1573	1.46755	5	1.44.7	7		3	1.46739	1.4737	1.4135	47.4.1	1.46730	1.44728	215			7	1	114.	1.44715	1.4.11	1.4713	114		-				£ 14.	1	1.1.15	1.4.	1.4.	1.172
C SPANITY	*		91194	1.46116	1.44116	1.44115	1.4115	1.46114	1.46114			1	1.4112	1.46111	1.41	1. :4187	1.46194	1.44100	1.464	1.4.41	7.5.E.	1.44.1	1.45074		1.4641		1.46847		X .	1.46818	1.46611	**	1.45497	1.45491	1.45945	1.45470	1.45473					1.45465	1.454.1	1.45439	1.65036	1.45431	-		27.6.			1.04413	1.45412	1.45919	3.45906	1.45.544	1,45988
SPECIFIC		2754-1	1.4372	1.43725	1.43725	1.41724	1.43722	1.4.1722	1.43721	1.4372		1.43714	1.43717	1.4 1716	1.43716	1,41714	13713	1.43714	1.43764	11760	1.4 1553	1.43645	7.27		***		1.443			135.45	1.4.357	1.4 7563	1. • 7553	1.47543	1.43533	***	1.4.7515			2072		1.43667	1.43.61	***	1.67.60	1-6 3662	15 4 7-1	1.44			7	1.4 7.00	10.4	10 % 4.1	1.43397	1.17%	1,((,,,)
			36.5	1-34619	1.3418	1.34617	1-30416	1.34-15	1.304.14	3.46.		1.34612	I. Jak 12	1.346.1	1.34412	L. 34-13	1.744.1	1.3421	1,344,38	1.3561	1.345.5	1-34013	1.35401	1.34710	1.3472	1-3476	1-74743	1.71.	1 20003	1. 1-413	1.30-72	1. 34. T	# 97	1.302-2	1.344.1	1.34.51	7		2000		7	1.30.07	I. Jana, R	1.38849	1.34476	1266.1	22	3.34072		1007	7	1.1.675	1.34.76	1. 34-70		1.34470	24:
		116911	1.14707	1.19363	1.137	56.61.1	1.19296	1.19743	1.19779	1.19276		10761	1-14755	1,14245	1.19243	1-14242	1.13761	1.19240	1.1.7.1	1-1-243	1.1975	1.19276	1.14.44	1-17365	1.19324	1.19%	161-1			25	1.19540	1.18.75	1.136.4	1.1365	1.13449	1.19715	M. 101.1		10101		1-10027	191-1	1.18%	1.136%	1-12077	3	- 1					1-10:00	22.7	. 1 23 %	1-13-14	1-124	1.7667
		•	:																!						ì																1																
			!																																																						
	•	/ See	2.00	15.9447	15.4437	13.96.26		15.04%	15.9983	15.24.2 15.84.2		15.477	15.4.5	15.4.6	15, 1784	15, 3941	15.2414	15.2262	15.1349	15,1716	15.1524	15.1175	15.1:34	15.1165	15.1057	15.6342	11.0016		12.6	15.0101	15.6461	15.6515	15.6571	15.0.21	15.0-1	15.844	12.9 27		700	CL 60 31	15.0141	15.01%	15.0121	15.09.7	15,4054	15.5526		-				14.5.72	16.49.41	14.21	14.77-5	: · · · ·	16.1794
	•		15,1717 15,9547		_		14.17.4 15.44.5	_	-	_				_	_	_	_	_	_		_		_	_	_	_	- (		•	1010101 10101	_	_	_	_	_	_							_	_	_	_	175			•	13.0113 14.013	_		-	_	_	_
11910	2	בו פולוילו	15.121	15.1715	_	15.1716	92/19/1	15.1781	15.1197	_	13-11-6		14.611	15.3	14.H978	14-92-3	14.7538	14.5444	16.4.57	1 01/5"	10.5726	16.4796	14.46[*	14.4977	14.3777	Z	14,374.		1000	16.2433	14.224	F-18-1	14.1433	11771	16.11.11	****	14.1277					10 157	14.4239	14-6104	13.9342	_	11.6775	13.6477	13.95.62		-	13,31,2	W. W. W.	4 - 4 - 6 :	13.7.5	_	13,756
ACTUAL WITHIE	2	CI 8121-41 FIG. 41	15.121	14.6326 15.1715 1	10317 15.1213	15.1718		14.42.90 15.1781	14.4292 15.1197 1	14.6245 15.1193	10,177	15.6476	12.5162 14.6113	1.0.a.	14.5461 15.H979	14.5-07 14.42-3	14,5150 14,7530	14,4749 14,5444	1-124 16.4.57	1 0165-01 14-5910 1	14. TE 14.572 ]	1-,7074 14,4794 1	14,2567 14,4414	14.216.0 14.4877	14.1739 14.3777 1	Z	14.6.45 14.37h.		10.01.0 10.01.0	16-2-31	11.90-4 14.2764	15.40.75 Lean 14.	13,6362 14,193	13,7049 10,1721	11.75 W 14.11	13,7247 14,134	13.0007 14.1777			1 5 5433 1 6437	11.5320 1	11,5355 14,035	13,4742 14,0239	11,2520 14,0108 1	11.6244 11.9362 1	~++5°E1	13.976. [ 14.676.	11.55 13.647	13,3364 13,9462		1 6119	11.7.1		4 - 4 - 6 :	13.11 " 132.m ]	1	1 - 0 - 0 - 0 - 0 - 0 - 0 - 0 - 0 - 0 -
	3 4 5	[	16.176 16.632 15.171 1	10,1752 14,6326 15,1715 1	16317 15-1213	THE THE PARTY OF THE PARTY OF		10-1764 14.6230 15.1201	19.11.71 14.6292 15.1197 1	10.1575 14.6285 15.1193	TOTAL TOTAL	14.57.37 15.61.00 1	18.16.7 14.516.71	10-1-1 1-0-0-1 11-1-01	16.1423 14,5461 14,8478	18.19.48 14.55.07 14.62.43	10.173 14.5150 14.730	16.144 14.4745 14.5844	16.1391 1-4.126 14.4.57 1	16,2107 14,3474 14,5718 1	14,2357 14,74 15,5226 1	12,7565 10,7076 16,6796 1	10713 14.2557 14.4414 1	18.c4.7 16.216# 16.6877 1	10,2449 14,1739 14,3777 1	10. 10. 10. 10. 10. 10. 10. 10. 10. 10.	15.25.4 14.4.4 14.37.		100 Mary 100 Miles 100 Mary 20 Miles		16.2447 11.90-4 16.2763	10-2-2-1 3-10-75 14-2-2-1	19.2331 13.6364 14.1433	16.2119 13.7049 10.1721	16.1441 11.75 36 16.1-14	19.10-1 11.7547 14.134	TACE TO LEGISTE CONTRACTOR			TA COLUMN TO CARD IN MARK	12.48.44 11.5320 1.54.64	11,5955 14,0457	\$.4559 13.6742 16.8239	9_4102 11_4528 14_8188 1	2000*[1 6429*[1 6954*6	11.4627 13.442	5. 45.6 13.374 13.4779	4.4325 13.3575 13.9477	13,3364 13,9462		1 2754 11 27512 1	Catholic 11 - 411		C-4-6: 605:11	1 4.135 13.1. " 132.m 1	1 6-2-51 2:48-11 12-5-6 I	Parall Banders Banke I

441	*****	# 15°	11.6123	17.73	14.763	1,20016	1	1.430	1.45447	1-46794	1.46016
37.	4 7 4 4	21	12.0744	13,6912	:	1,200%	. 3000	1.43387	1,45467	1,46705	1.46916
4.3372	**	****	12,0375	13,6462	14.67%	1.20631		1,43384	1,4598	20134.1	1.4431
£ 22.75	1.74.4	1917	17.9422	13,6400	14.4787	1.20 X		1°4 1383	1.55	1,44705	1.46417
5.28TS	4.355	377	12.94.4	13.478A	:. ::	1,20645	7	1,43361	1,454	1.447#5	1.46417
5.2772	4.32mg	4. yees	12,4377	13,603	14.463	1.20051	L. X	1.41379	1.4546	1,46705	1.44917
25.5	4. W45	. ×	15-44-21	13,544	10.6506	1.26%		1,43378	1.45464	1.44705	1,46917
کرر ۲۰۰۶ م	9.5729	1675.0	12.77.20	33.5746	18.6543	1.2863	- X - X	1.43777	****	1.4175	1.44417
1.2155	1592.0	44.2°5	12.7592	13.541	16.4523	1.200%	A. Mark	1,43376	1.45424	1.46705	1.46417
5.1 Xe	4.2175	4.2843	17.73	13.5468	14,6587	1-28557	-	1,43374	7.5.5.	2.47.5	1.46917
1-174	4.1462	Į.	12.7146	13.5322	14.45	1.20040	 	1-43373	7.5.5	1,44785	1.46017
3	4.1636	9.15il	12,7013	13.43	7.F.S	1.20071	I. Med 7	1.43372	1,45985	1.44785	1.40417
6-1337	21117	121.	12.6454	13.52	21.1	1,20672	J. Yanna	1.43372	1.45905	1.44785	1.44917
6-11.34	4.1117	4.8482	12.4729	11.5	1.47	1.26672	1. Jeens	1.43171	1-45005	1.44705	1.4-417
27.1.4	9.BA71	5.0 PS 7	16.05.5	13.5754	10.00ts	1,28672	- 35 FEB.	1.43378	2.45465	1.46705	1.4417
15/90	1 12.	£55.	12.04.93	13.513	10.0462	1-20072		1.43%	7.45005	. 57.53	1.46917
5-8505	4.6-69	4.03¥	151 v-21	11.52	7.1.1	1.20670	1.34:00	1.413%	2.45.00	1.46785	1.46917
6-C 3+5	1010.	4.23	12.22	13.5777	**:	1.20099	1. Xer.	1.43347	1,45005	1,44785	1.66417
5-82r3	7	0.4.70	12.629-	13.5.57	15.44.41	1-20057	1.74	1.43347	1.55	1.4785	1.45917
E-8762	A 20.4	5	12.4127	3.5238	14,844	1.20055	1. Mr. 0	1.43366	*****	1,4735	1.46917
3.5	2.855	124	25.0m.21	13.>222	14,0445	1.20052	1.384	1,43345	1,556	1,44785	1.45.917
5,4727	21.7	- X.	12.74.4	3.5725	14,0442	1-20650	S. Jakes	1,43364	1.45964	1.44765	1.46917
5	4.627	B. +3357	12.5427	13.5192	***	1.26%	1. year	1.42363	1.656	1.44705	7.53.1
2.4.3	# 10. T	4205	12.540	11.5178		25387.1	- X	1.43763	1.45.4	3.06705	1.44917
5.=355	***	A. 4151	12,5414	13,5145	****	1,20053	Serie To M	1,43352	1.454	1.44785	7.649.1
5-4177	1244.8	***	12,5753	13,5153	16.4.31	1.20050	- 344 C	1.43361	1.45463	1,475	1.4.17
5.4275	To the W	14.4	112-5714	13.5142	27.1	1-2004	7.75	1.43%4	1.5583	-, tit	1.45.917
5.82-8	1.4.4	A. 4.	12.54	13,5131	14.427	1-7866	. H.	1.43366	1,45907	1.44765	1.44917
IL Mary	#548"P	8_6761	17.52	13,5121		1.706	L. Marke	1.43359	1.4590]	1.44785	1.46417
210.5	144.4	6.8675	12.5542	1115-11	14,4421	7.2062	1.3604.1	1.43754	1,45463	1.44785	1.44917
2.4.51	4.4277	1.5.4	25.5.21	17.5162	15.4.21	19802-1		1,4176	1,4541	1.44785	1.46917
5.5.17	1714.4		12.54	13.5043	14.41	1.20639	- X	1.43357	1.45463	2.42.5	1.44917
(1)	2.44	4	12.546	7.23	14.41	#3C.1	- X	1.43 757	1.454.1	- 1175	1.44.917
5.4.3.7	1.77.	A.A374	15.5.71	11.5677	14.41	1.75%	1.3441	1	1.454.1	1.475	1,44917
5.4275	A.7-74	6.4313	17.546	13.5676	14,415	1.200.17		1.433955	1.45403	1,44785	1.5517
2-41-12	8. Te92	P. H.	17.5 164	13.5004	14.41	1.20037	1.35401	1.43355	1.4541	1.47.15	1.46917
5123	4.11.4	# 1 F '8	12.5136	11.4457		1.20637	<u> </u>	1,43355	1.45.43	1.45785	1.44917
5.465	10-7-61	4,-113	12.5 344	11.565	14.41	1.20637	7	1.4335	1.45403	1.44785	1.46917
2-13-5	27.	1166.	12.5242	13.5447	14.41	1.28637	1. year.	1.43754	1.45483	1.475	1.46917
5. 7926	#. 75.75	+-852c	15.5%	13.54.2	15.41	1.20637	1-3446	1.43354	1,45403	1,44785	1.44917
5.74.9	8.7476	. 7973	12.5231	X X E	14.71	X92.1	1.30092	1.+3353	1.45983	1.44785	1.46617
5. 7m58	6.7.23	CZ.	12.5284	7.23	2112	1.2639	-3445	1.43353	1.5543	1.44705	1.46917

| 1440| | 1400| | 1750| | 2700| | 2710| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700| | 2700

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.27374	.27374	27176	.27374	.27374	27374	ALC 12.	27776	. TTT.	20175.	.7572B	£.	Ç.	i k	N. A.	X view	.XX.	212	Ç.X	***	27.515	X		25.55	\$ \$ \$ \$ \$ \$ \$ \$ \$ \$ \$ \$ \$ \$ \$ \$ \$ \$ \$	i X	X	2552	75517		**		K	X.			¥.			ž,	23610			Ž.		Ć X	X 537	*	142.
.7.000		76.62	24620	7.087	7		7	26006	SACA!	.7613E	× 4.4.	1 K	Ĉ.	25.53	2	£17.	-2-666	24.06.7	757.25	-2000	\$ <b>4</b>	152	2000	26432	24.4	2.333	5 K.	24.274		24145	24145	7112	10074	7.066	7	26.25	-23964	2726.7	.2338	-23e14	2	2387	23857		77.6	24.4	23433	23549
• 2.	1946.		3.4	-74462	19467		7	75457	\$5.×	25-92-	24.62	71.35		26.23	76.143	336.	X .	Ç X	X	29.54	* * * * * * * * * * * * * * * * * * *		7.2	2514	, X	14542	250.5	25.33	26.96	****	7,547	7.44	.74471	26.3%	22.2	50.22	3.	X 100 X	7.0%	610m2°		23917	23806	23426	276	X 36.2	23575	.23511
5 -20016	5192.	2001	.20c17	29915	. 26913	1100%	1000	10002	# 1.	\$ <del>4</del>	1,4003		997		2000	*29C1	**	7	-2e Brit	******	20000		24076	24055		1.47	fiet l'			A.7.	21761-	117	evs.1.	15421		-1436	1933	1404	19291	.19163	2161	1	11961.	7	75071	24.61	.19733	116672
* X. 34.	K.	26.75	187 A	CAR.	£.	7437		78.87	38.5%	.74.961	24.35A	٠٠. ١	75.176	26.463	27.5	PI - 42°	74147	-24[5]	24949	4	15.X	֓֞֞֜֓֓֓֓֓֓֓֓֓֓֓֓֓֓֓֓֓֓֓֓֓֡֓֓֓֡֓֡֓֓֓֡֓֞֓֓֡֡֡֓֡֓֡֡֡֓֡֓֡֡֡֓֡֡֡֡֓֡֓֡֡֡֓֡֡֡֡֡֡	XX	8	X X	17.7%	3,4	X .	i X	X.X	7	7. T.	*X**	24.5	15372	74.803	¥. 2.	1777	24.7	*56384	***		200	24370	54.74		2012	54845
3.2255	-21216		15112	.21136	22112-			21879	.21070	.21951	710%	71047	7.416	2132A	.21623	2017.	.21917	21016	71017	F5815.	.21927	7.517.	71937	-21637	7167	61416	-21804	\$ 5 P. S.	13.5	15065	V 1. X 1. V	54.762	Ph 149	237.1	2007	.26631	2020	265.30	786.4	28424	2. X	3116	.28281	7.7.		2012	2067	20039
2621.	1501	15.757	15731	15704	5		¥ 1	15622	116.11	15630	15541	15543	25.55	3	15551	.15552	9.45.	1-551	1555	14579	1457		6195	-15+3a	15659	3.351	51721.	25725	1,7,51	1-7-1	Z.:	61721.	.15783	.15-43	2	£995] ·	1557	15.40	19461	15451.	15.001	1630	15259	15210	51 / T	<b>9</b> 351	95.75	7
.17976		776	178.3	.: 7423	-1 refe	2777	2777	17750	17751	.17743	\$1775	427.1	277.1	17711	177: 7	-177:4	-177.1	-177.	51715	.1777-	1771	1776.	17751	.177.55	#1-14 #1-14	\$/4¢	2.17.	17435	1176	17-54	* 12 L T		717714	2 4 4 4	10.00	17571	.1757a	57.6	17499	.17364	.17372	17236	17193	17157	111.1.	1/0/1		44.00

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1.84625	1.84645	1.054.1	1.85216	1.65375	1.85521	1.45657	1.85786	1.85908	1-84027	1.A6164	1.96258	1.86373	1.85486	1.86599	1.86712	1.86.823	1.86934	1.87042	1.87148	1.87252	1.87352	1.97448	1.87541	1.87630	1.47714	1.A7796	1.87869	1.87940	1.88004	1.84068	1.Re125	1.94178	1.84226	1.8827	1.RA 316	1.94346	1.9837R	1.88407	1.68431	1.88452	1.88469
5.64926	5,65543	5.05156	5.04753	5.07337	5.07905	5.CA456	5.69990	5.04510	5.10014	5.10506	5,10984	5.11.54	5.11910	5,12356	5.12791	5.13214	5,13626	5.1.027	5.14616	5,14793	5,15159	5.15514	5.15857	5,15149	5,16510	5,15820	5.17120	5.17410	5.17690	5.175.0	5,18241	5.18473	5,16717	5.1.351	5.19179	5,19395	5.19405	5.19409	5.20004	5,26191	5,20371
5.56198	5.49825	5.49468	5.49124	5,46743	5.48477	5.48177	5.47492	5.47425	5.47373	5.47137	5,46918	5.44712	5.46521	5,40343	5.46177	5.46027	5,45877	5.45740	S.45n12	2,45,402	5.45340	5.45273	5.45172	5.45078	5.443HG	90657.5	5.44927	5,44754	5.44686	5.44623	5.44554	5.44511	5.44462	5,44419	5.44179	5.64365	5,44315	5.44749	892775	5,44251	5.44238
6.67031	6.44644	4654596	6.45983	6.45764	6.KS45A	6.45237	440544	6.64×75	6.64725	6.44547	6.44474	5.44367	6.44269	6.64178	5.F4047	6.64010	6.43929	6.47HSB	6.5.177	6.63596	51414.9	6.4353A	14464.6	6.43394	6.43304	6.47234	6-63162	6.43092	6.4.1025	9.4234B	6.47900	6.47844	4.42792	5.47745	40754.8	6.42464	6.62536	6.42410	6.6254	5.42575	6.42567
70626-02	20.89502	20.88142	20,84835	26.85593	20.84416	20,43321	20.R2311	20,41302	27.40570	20.79H4B	PC.797.95	26-7-710	20.74.243	20-11-12	20,11743	70.17547	20.7752A	20.17525	20,17579	20.17.73	20.77-15	20.77978	2C. 7H1-6	20.74350	29.7×543	20.7A711	20,78939	50,79073	20.7021A	20.74343	20.7-4-5	29.73522	30,74576	20.75404	20,795.09	20,75550	20.7954R	20.79624	26,79403	20.75302	20.79164
2,13916	2,33915	2,33916	7.33917	2,33918	2,33914	2,33419	7,33420	2,33929	2,33~20	15016.5	15-11-5	15-18-7	15.51.5	170iE 2	2,31471	2,33421	2,33420	2,31~20	2.33-29	2.33+20	2,33.420	2.33420	2,33919	2,31419	2,33419	2,33419	2,33919	2,33419	5,33919	2,33419	2,33519	7,33919	2,33919	2,33519	2,33913	2,33919	5,11919	7,33519	2-33-20	7,33320	2,33920
22.62564	22.63.21	22.54169	22.54RG6	22.65327	22.65741	22.65081	66244.55	25.45.52	22.6,14.55	- 60+06-22	27.64290	22.55114	22.55493	66.45.33	7.4634.	15000022	647.47.53	22.5-451	22.5.1A3	2 A 3+2ª	22.6.3.42	87.6 7.5	27.43315	22.4 1176	176.5.37	44674.25	77.42.41	22.52.55	97.52.55	27.45933	27.43113	22.53213	22.433.5	50-64-05	22.63459	22.5.3000	77.54335	22,64244	27.4-463	22.54.599	22.64626

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1 7 10.	•	97610	•	2410.	•	. 1755				•	•	•	•		• •	•	•	•	67619	•	•	•	•	•	•	10. ALICA.	• •	•	•	•	10/20	• •	•	4 KKK 6 C	•	•	05.30	• •	•	•	•	7550	•	7	7	12563	•	•		•
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5.24281	25308	.26310	.26317	24323	156.327	561.45	20.00	24.34.1	24345	24.368	24350	. 24 45	24345	5	95.46	26347	-243A2	*0-u2*	20.00	50947	26.737	34545	E44.20	\$1542°	625 +2	12542	14542	4 - J 4 - 7 - 8	.26513	4505.	25.44	24.63	50000	26618	24343	-2537P	26362	96197	.26321	-26309	26293	12757	26266	.26259	.26.52	26244	26.246	26222	26215	26207
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0.726*	. 894 1	.86863	67470	.26165	.11670	.62635	. 86572
*5726*	61463	. A6803	45474	.24145	.11647	.02537	
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34121	63760	.H6+29	.45577	.26199	11863	21920.	. 6657
.72179	60750*	.85427	.45541	-26/95	.11ag	.42674	. 88574
.7150.	60460*	15478.	585,37	-26205	.11965	.12676	.005A
61120*	\$ 07.0°	H542H.	.4558A	*2626A	\$1611.	. 62678	.60586
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44155	60450	16+98*	*4554*	.26213	5:611.	.02601	. 885.
5412c"	£0760°	. 46433	96557*	.24216	.11943	.076A3	.00581
A-1:0.	69750	.85435	16557"	\$12.5	11952	.02684	.06581
PA155.	63760	£4436	66557*	.26220	.11%	.026A5	.605.
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54175	60760	8643B	. 55401	.24223	.11975	.07688	.40542
6×120°	60700	.BF438	10057	*5555*	29911	326.9	.0058
96126	60-60	.84437	20457	542.45	.119PA	.02689	.09562
20120*	60760	.B6436	.45603	.25227	:1845	96929*	.00562
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93//	60750	.84431	*45604	.24229	90021"	16420.	. 80542
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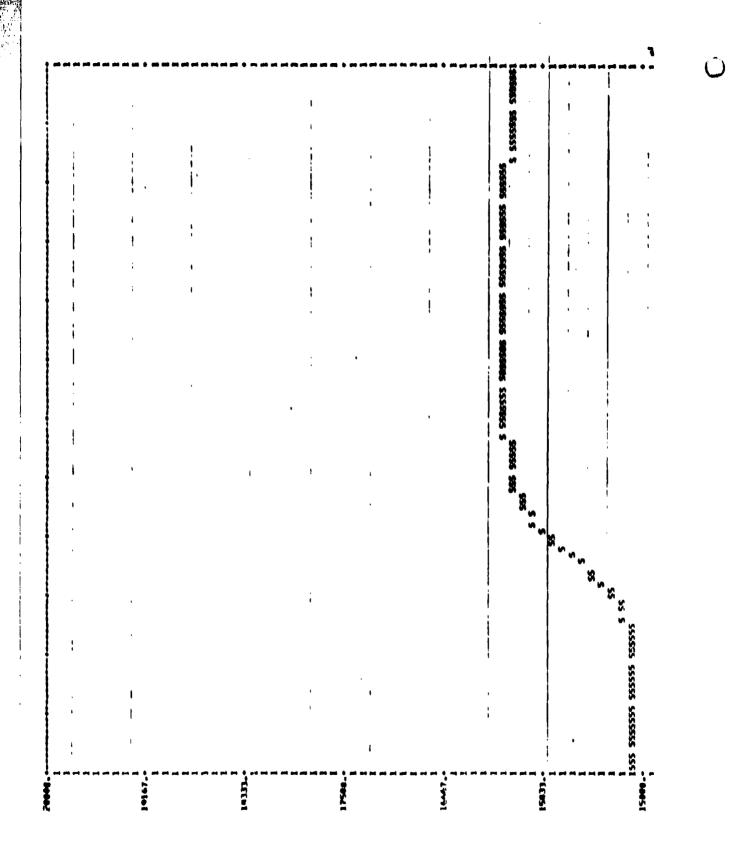
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## APPENDIX C

Dynamic Simulation--Program Flowchart

Degin

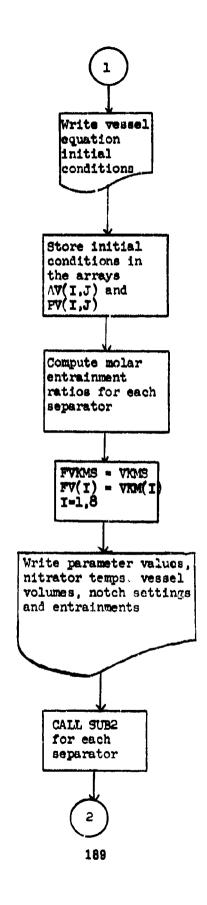
Read values of frequency factors and activation energies in rate equations

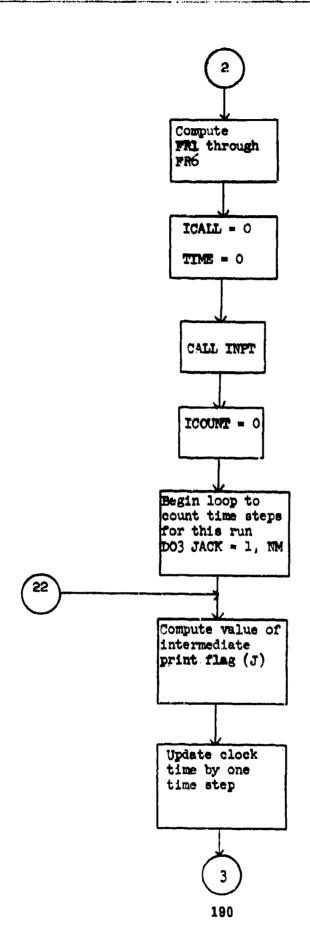
Read values of other constants and parameters in vessel equations

Read values for heats of reaction, densities and specific heats

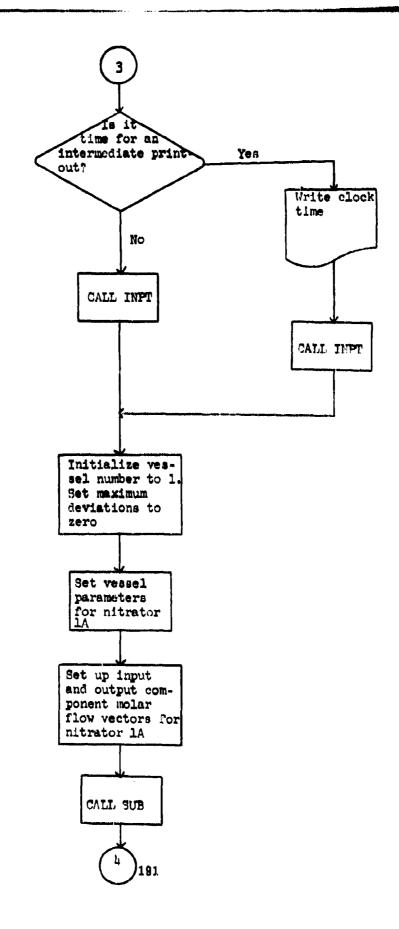
Read initial values of molar component flow rate for each vessel, i.e., initial conditions for vessel equations

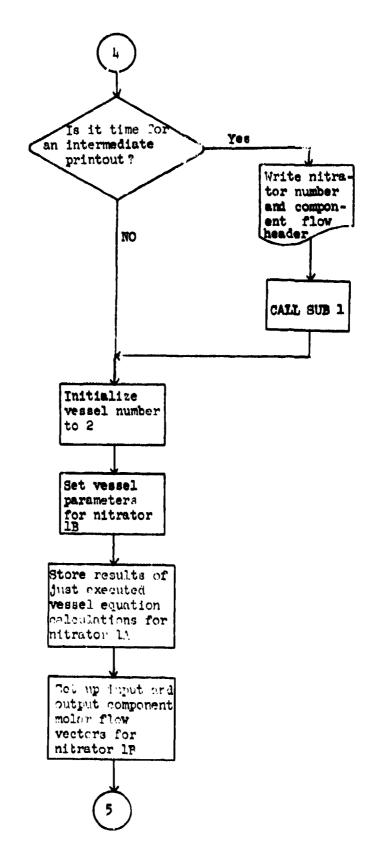
Read notch settings, entrainments, vessel volumes nitrator temperatures and EX

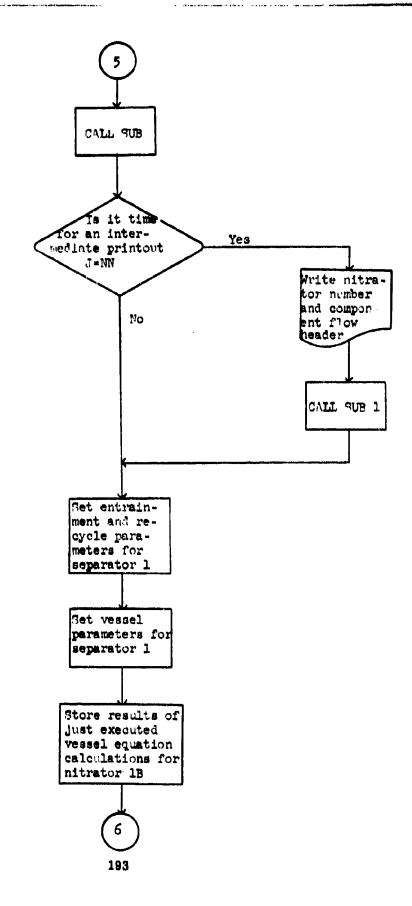


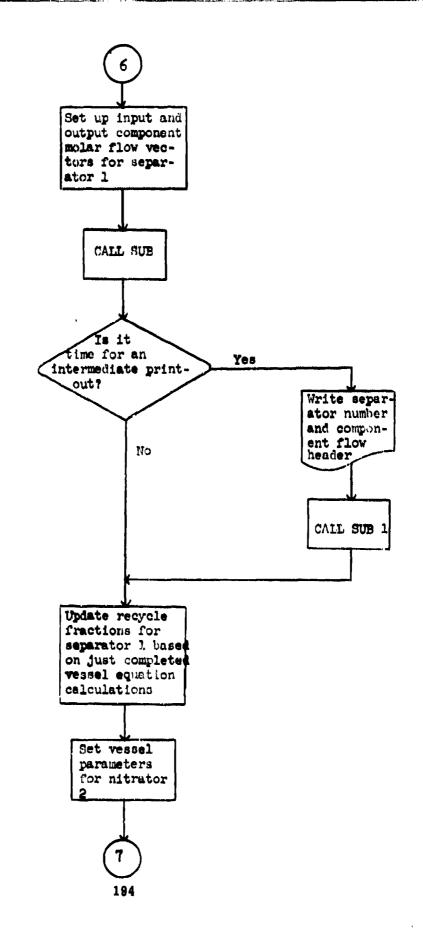


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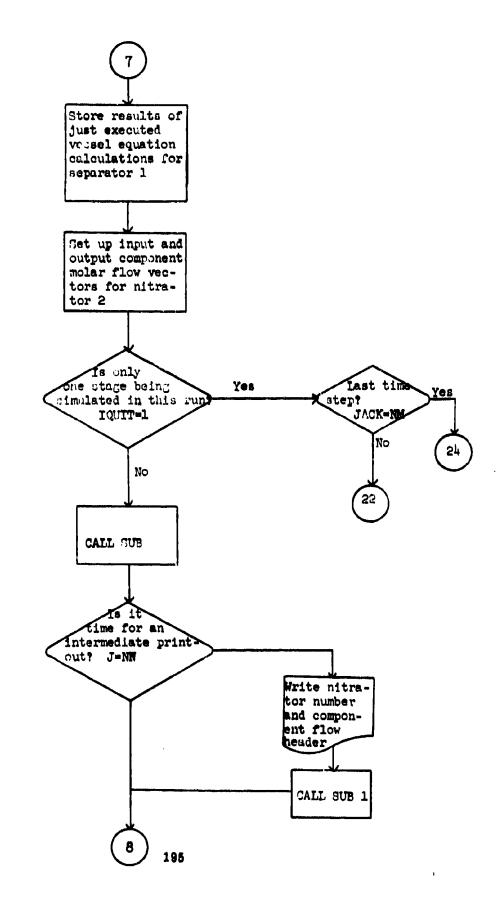




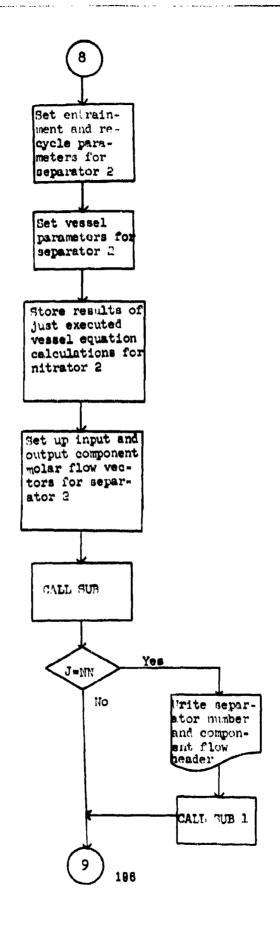


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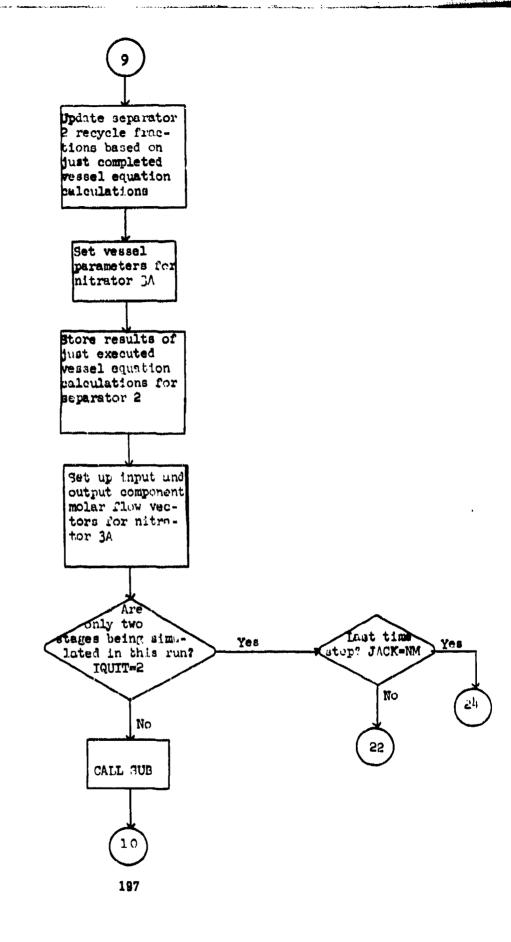
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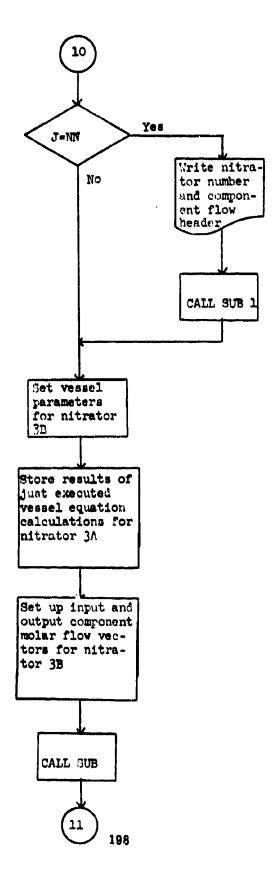
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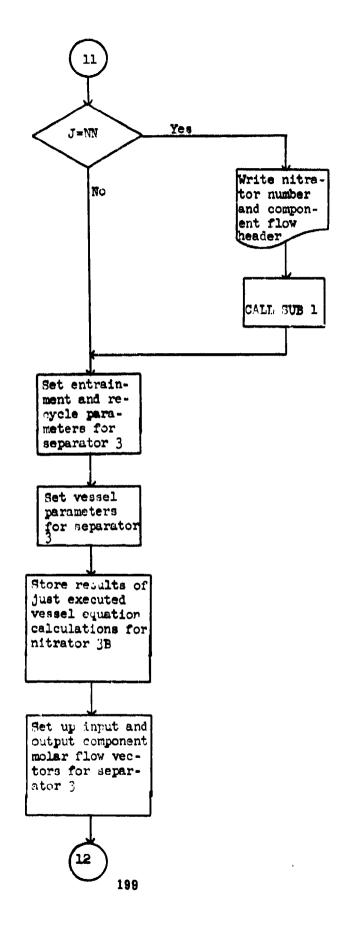
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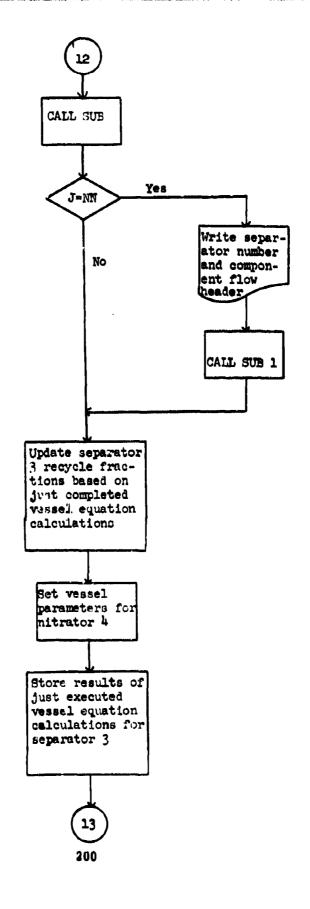


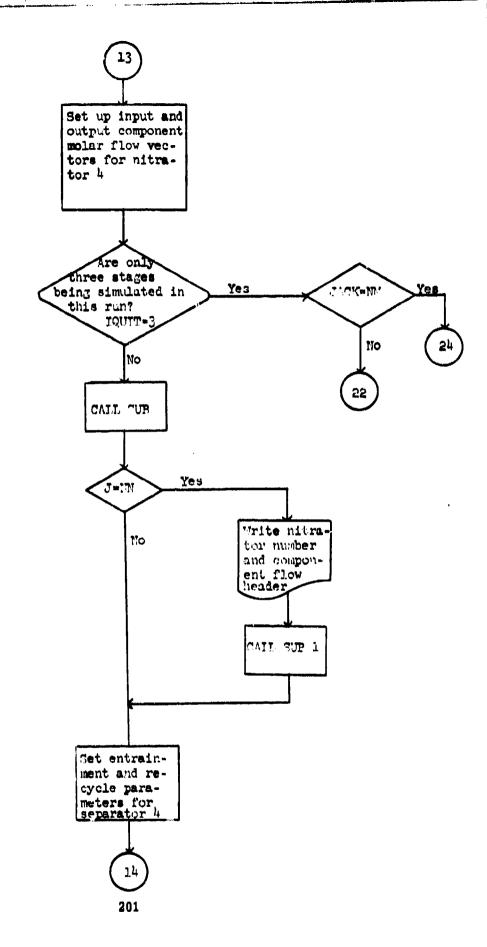
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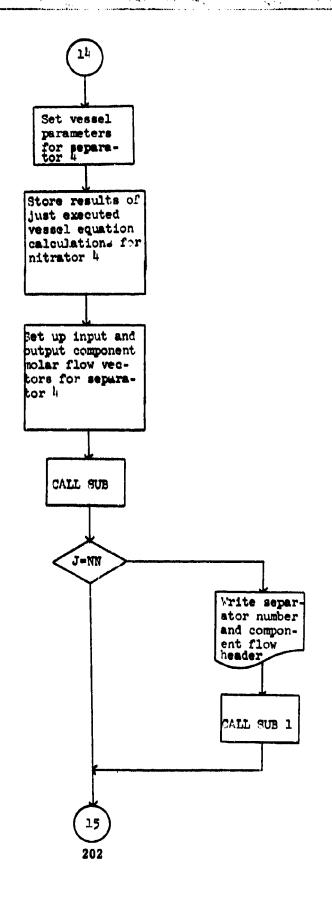
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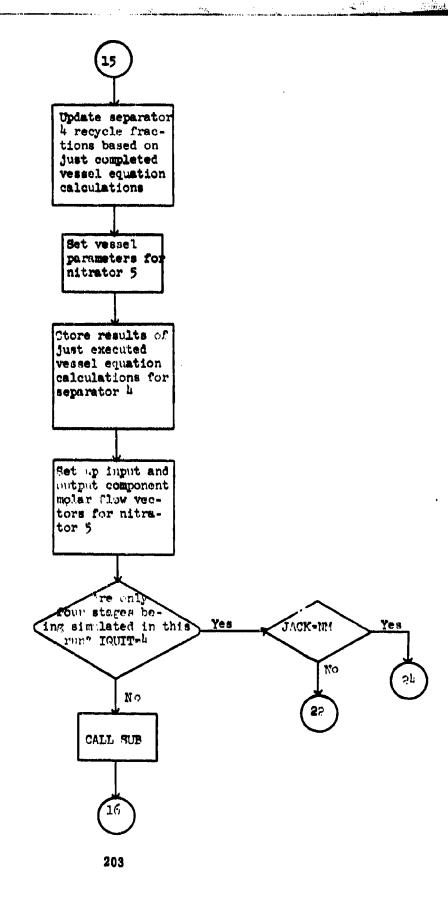


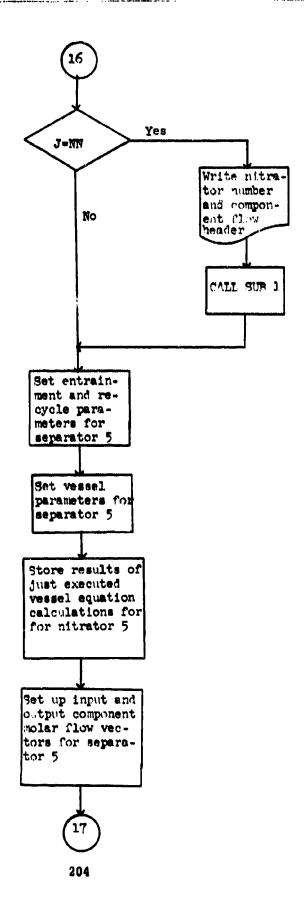


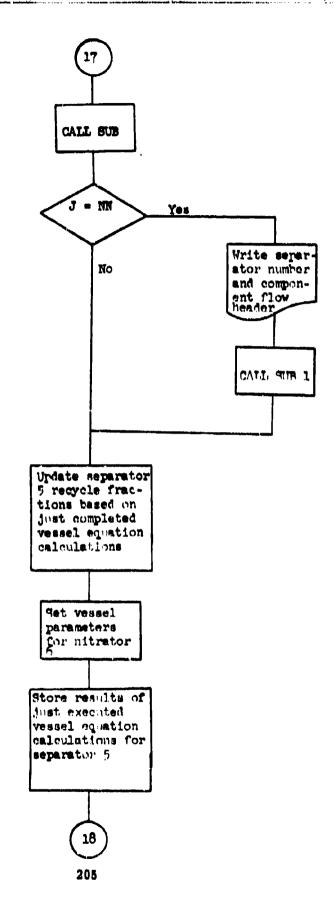


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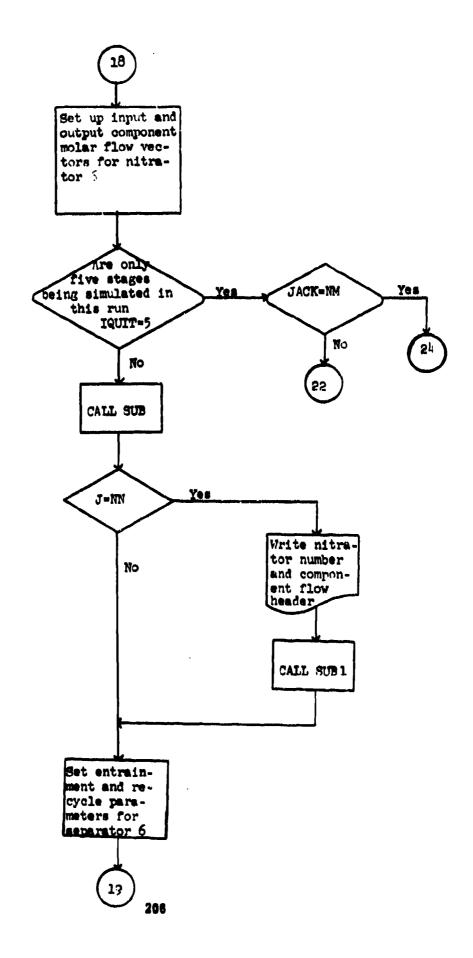






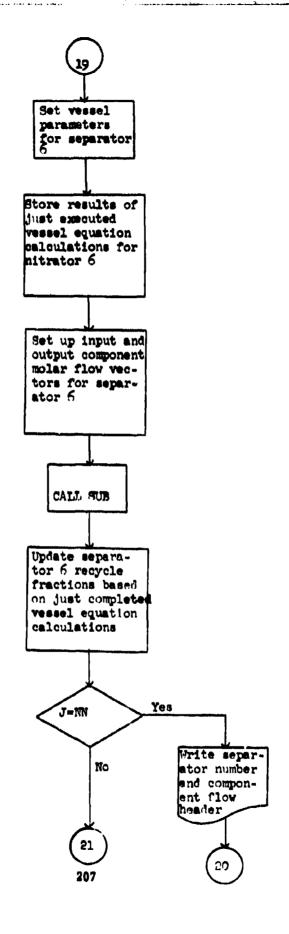
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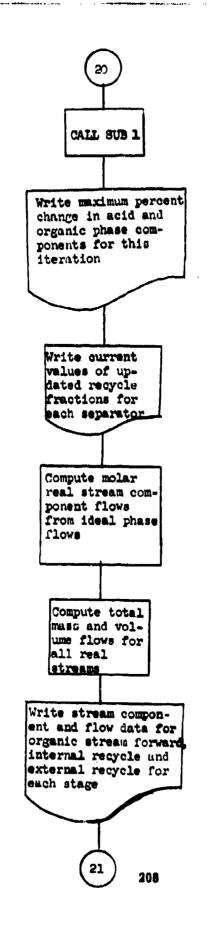


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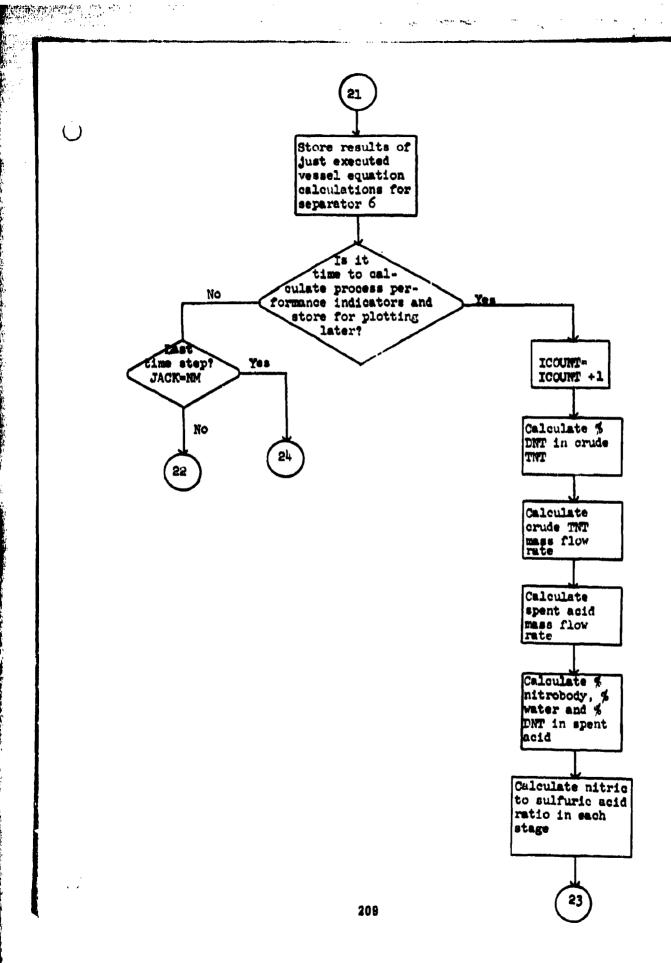
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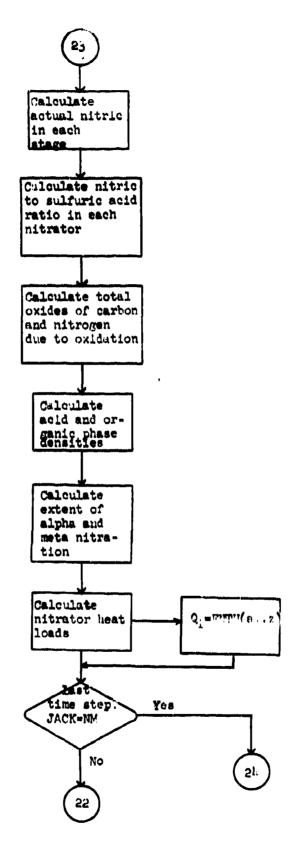
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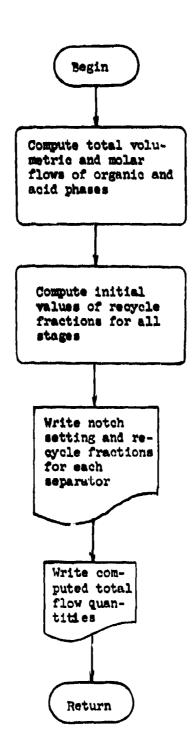
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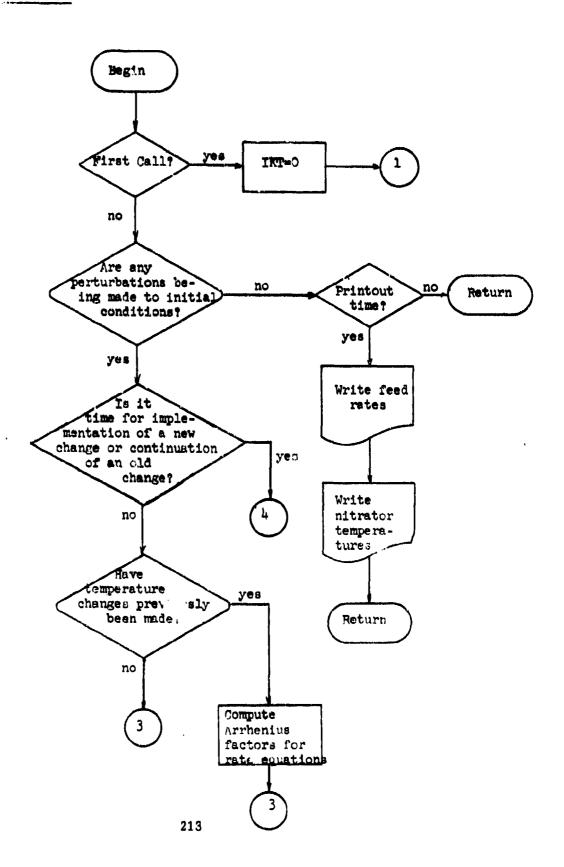


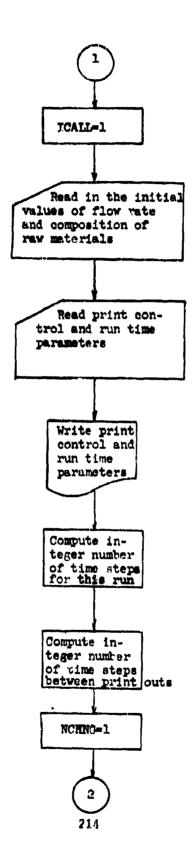
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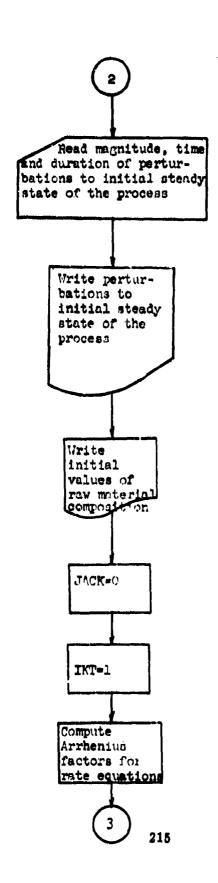




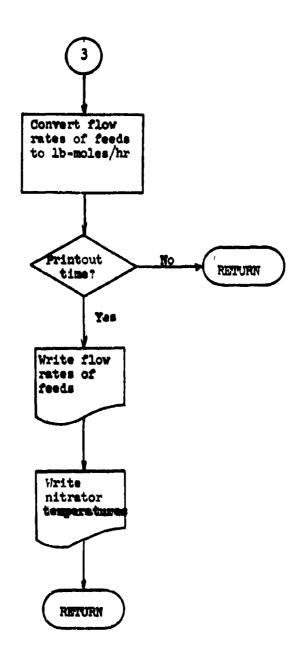


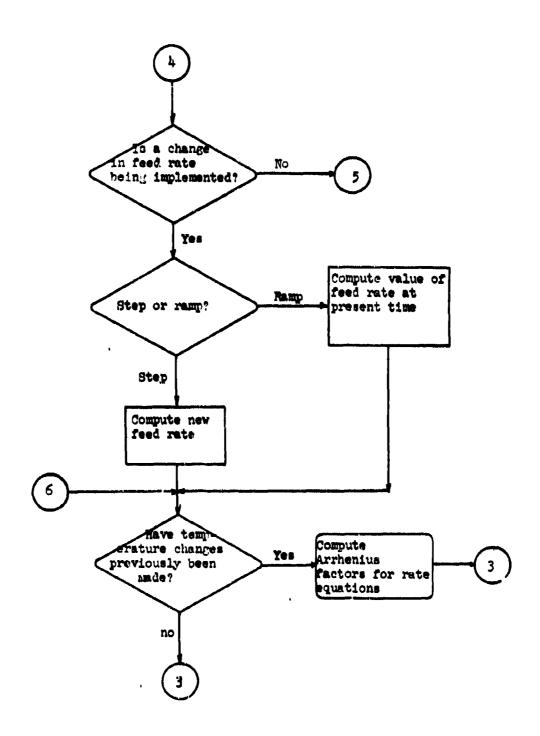


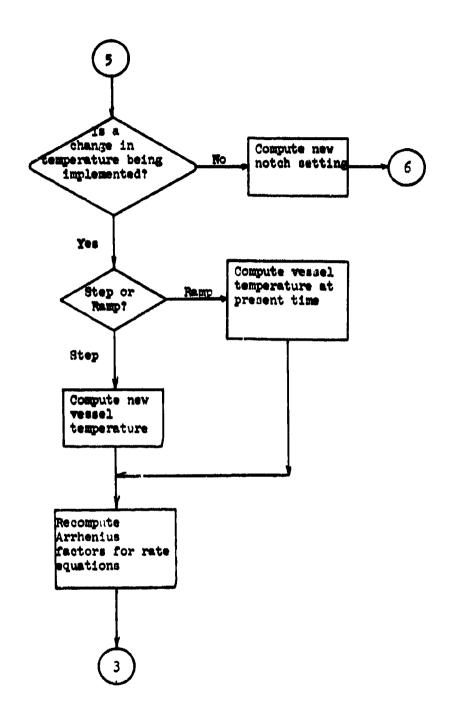




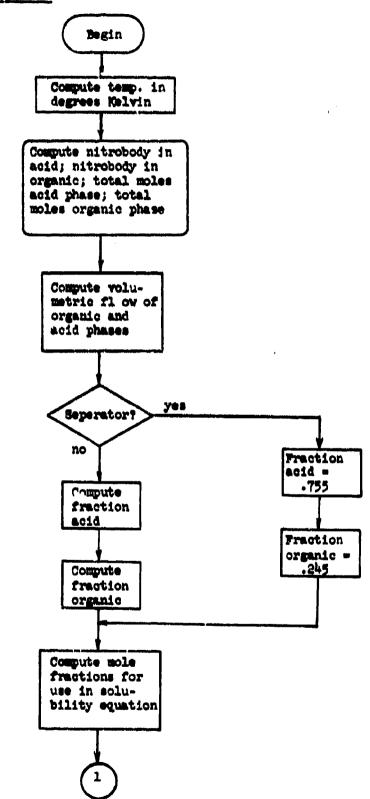
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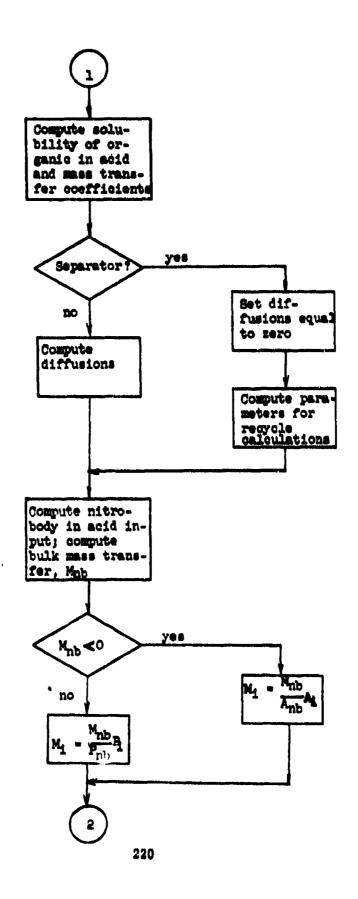




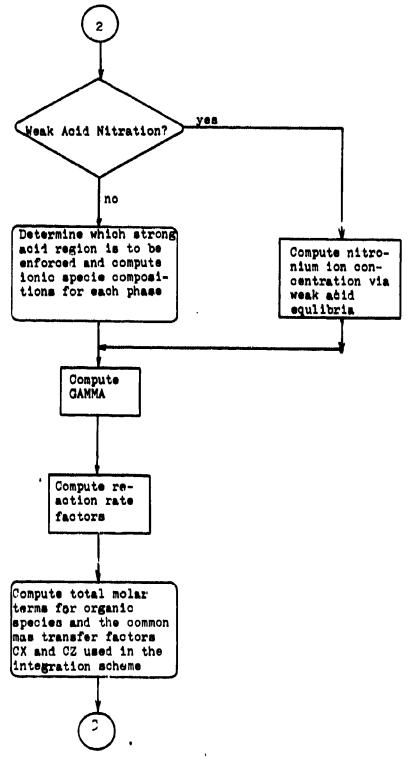


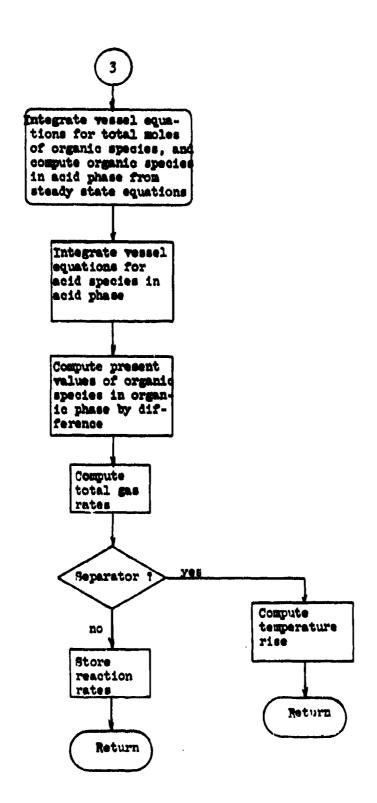
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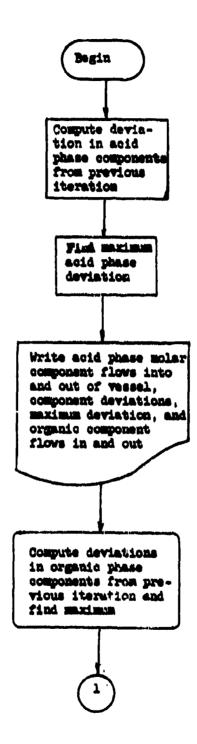




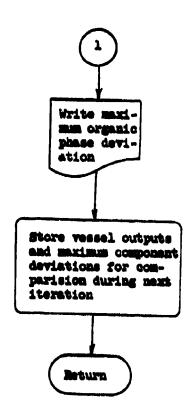
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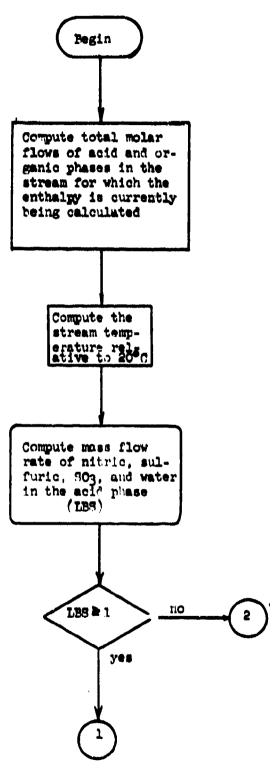




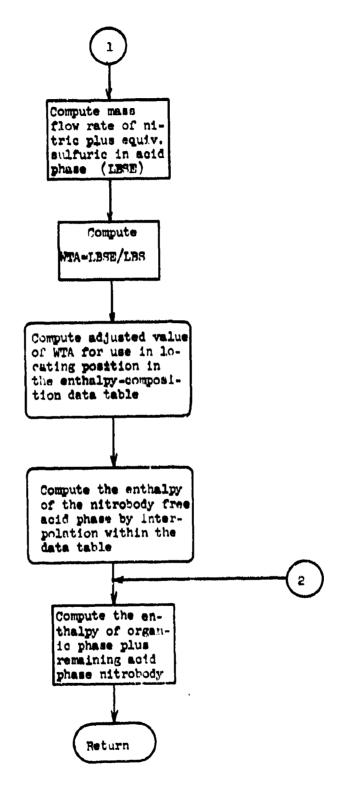


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## APPENDIX D

Dynamic Simulation--Program Nomemclature

Main Program	·
A (I)	moles/fir of component i in the total acid phase leaving the vessel currently being calculated
AA (I,J)	moles/hr of component i in the stream phase j of the separator currently being calculated
AF (I)	moles/hr of component i in the acid phase of the organic stream leaving a separator
AI (I)	moles/hr of component i in the total acid phase input to the vessel currently being calculated
AIR (I)	moles/hr of component i in the acid phase of the internal recycle leaving a separator
AKMA	acid phase mass transfer coefficient
AIC10	rate constant for reaction 10
AK12	ratio of rate constants for reactions 1 and 2

AK4S rate constant for reaction 4 in strong acid

AK4W rate constant for reaction 4 in weak acid. NOT

rate constant for reaction 3 in strong acid

rate constant for reaction 3 in weak acid. NOT

USED

USED

AK3S

**AK3W** 

AK5S rate constant for reaction 5 in strong acid

AK6S rate constant for reaction 8 in strong acid

AK7 rate constant for reaction 7

AK8 rate constant for reaction 8

AK9	NOT USED
AN (I, J)	percent actual nitric acid in the acid phase of the external recycle leaving separator j at the end of the ith plot interval
ANA (I, J)	amount of $\alpha$ - nitration taking place in nitrator $j$ at the end of the $i^{th}$ plot interval
ANAC	running sum used in computation of ANA(I,J)
ANM (I, J)	amount of m-nitration taking place in nitrator j at the end of the ith plot interval
ANMC	running sum used in computation of ANM (I,J)
ANT (I, J)	moles/hr component i in the total acid phase leaving nitrator j
ARRAY(I,J)	moles/hr of component i from fresh acid feeds to nitrator j
AS (I,J)	moles/hr of component i in the total acid phase leaving separator j
ATM (I)	total moles/hr of stream phase i leaving the separator currently being calculated
ATW (I)	total lb/hr of stream phase i leaving the separator currently being calculated
AV (I,J)	initial value of moles/hr component j in the total acid phase leaving vessel i
AVL (I)	total cu ft/hr of stream phase i leaving the separator currently being calculated
AXR (I)	moles/hr of component i in the acid phase of the external recycle leaving a separator
A1A (I)	moles/hr of component i in the acid phase leaving nitrator 1A

A1AI (I)	moles/hr of component i in the fresh acid feeds to nitrator 1A
A1B (I)	moles/hr of component i in the acid phase leaving nitrator 1B
A1BI (I)	moles/hr of component i to nitrator 1B from fresh acid feeds
A18 (I)	moles/hr of component i in the total acid phase leaving separator 1
A2 (I)	moles/hr of component i in the acid phase leaving nitrator 2
A2I (I)	moles/hr of component i to nitrator 2 from fresh acid feeds
A28 (I)	moles/hr of component i in the total acid phase leaving separator 2
A3A (I)	moles/hr of component i in the acid phase leaving nitrator 3A
A3AI (I)	moles/hr of component i to nitrator 3A from fresh acid feeds
A3B (I)	moles/hr of component i in the acid phase leaving nitrator 3B
A3BI (I)	moles/hr of component i to nitrator 3B from fresh acid feeds
A3S (I)	moles/hr of component i in total acid phase leaving separator 3
A4 (I)	moles/hr of component i in acid phase leaving nitrator 4
A4S (I)	moles/hr of component i in total acid phase leaving separator 4
A4I (I)	moles/hr of component i to nitrator 4 from fresh acid feeds

A5 (I)	moles/hr of component i in acid phase leaving nitrator 5
A5I (1)	moles/hr of component i to nitrator 5 from fresh acid feeds
A58 (I)	moles/hr of component i in the total acid phase leaving separator 5
A6 (I)	moles/hr of component $i$ in the acid phase leaving nitrator $\theta$
A6I (I)	moles/hr of component i to nitrator 6 from fresh acid feeds
A68 (I)	moles/hr of component i in the total acid phase from separator 6
BASIS	NOT USED
CDNT	weight & DNT in crude TNT leaving separator 6
CDNTSA	weight % DNT in the nitrobody dissolved in the spent acid
CHAMXØ	NOT USED
СНРМХФ	NOT USED
CNBSA	weight $%$ nitrobody dissolved in the acid phase of the spent acid leaving separator 1
CP (I)	specific heat of component i
CRUDE	lb/hr crude TNT leaving separator 6 (i.e., the mass flow of the total organic stream leaving separator 6)
CWSA	weight % water in the acid phase of the spent acid leaving separator 1

DENS (I,J)	specific gravity of the organic phase of any of the streams leaving the j <sup>th</sup> separator at the end of the i <sup>th</sup> plot interval
DENSA (I,J)	specific gravity of the acid phase in any of the streams leaving the j <sup>th</sup> separator at the end of the i <sup>th</sup> plot interval
DFAMXØ	maximum & change in an acid phase component between two successive iterations of the nitration section
DFPMXØ	maximum & change in an organic phase component between two successive iterations of the nitration section
DH(I)	heat of reaction for the ith reaction in BTU/lb-mole of reactant
DLT	estimated temperature of entrained organic phase that would be returned to nitrator 1B from the after separator
DT	itegration time step (hrs)
DTPR	time between plotted points (mins)
BA <b>Q</b> (I)	molar ratio of acid phase in the organic stream to total organic stream leaving the ith separator
EAQP (I)	molar ratio of acid to organic phase in the organic stream leaving the ith separator
EAQP1	equivalent to EAOP (1)
BAGP2	equivalent to EAOP(2)
EAØP3	equivalent to EAOP(3)
EAQP4	equivalent to EAOP(4)

equivalent to EAOP (5)

EAQP5

0	EAGP6	equivalent to EAOP (6)
	ЕЛР	value of EAOP(I) for the separator currently being calculated
	epr	enthalpy of mixing for fume recovery acid
	EQA(I)	molar ratio of organic phase in the external recycle to total external recycle leaving the separator i
	EQAP (I)	molar ratio of organic to acid phase in either of the acid recycle streams leaving the ith separator
	EQAP1	equivalent to BOAP (1)
	EQAP2	equivalent to EOAP(2)
	e <b>g</b> ap3	equivalent to EOAP(3)
	egap4	equivalent to EOAP (4)
	EGAP5	equivalent to EOAP (5)
	EQAP6	equivalent to EOAP(6)
	egl Egp	enthalpy of mixing for 40% oleum value of EOAP (I) for the separator currently being calculated
	EWN	enthalpy of mixing for weak nitric acid
	EX	imposed upper bound on the Arrhenius co- efficient in the reaction rate expressions for reactions 3 and 4 (MNT to DNT)
	EYW	enthalpy of mixing for yellow water

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**B10** 

E21

activation energy for reaction 10

ratio of activation energies for reactions 2 and 1

E3S	activation energy for reaction 3 in a strong acid medium
B3W	NOT USED
E48	activation energy for reaction 4 in a strong acid medium
B4W	NOT USED
E58	activation energy for reaction 5 in a strong acid medium
E68	activation energy for reaction 6 in a strong acid medium
E7	activation energy for reaction 7
E8	activation energy for reaction 8
E9	NOT USED
FA(I)	rctio of acid phase leaving ith separator that is recycled to total acid phase leaving the ith separator (fraction of acid phase recycled)
FA(I) FACID(I)	recycled to total acid phase leaving the ith
	recycled to total acid phase leaving the ith separator (fraction of acid phase recycled)  ratio of acid phase leaving the ith separator in the organic stream forward to total acid phase leaving the ith separator (fraction of acid phase
FACID (I)	recycled to total acid phase leaving the ith separator (fraction of acid phase recycled)  ratio of acid phase leaving the ith separator in the organic stream forward to total acid phase leaving the i <sup>th</sup> separator (fraction of acid phase sent forward)
FACID(I)	recycled to total acid phase leaving the ith separator (fraction of acid phase recycled)  ratio of acid phase leaving the ith separator in the organic stream forward to total acid phase leaving the ith separator (fraction of acid phase sent forward)  equivalent to FACID(1)
FACID1 FACID2	recycled to total acid phase leaving the ith separator (fraction of acid phase recycled)  ratio of acid phase leaving the ith separator in the organic stream forward to total acid phase leaving the ith separator (fraction of acid phase sent forward)  equivalent to FACID(1)  equivalent to FACID(2)
FACID (I)  FACID1  FACID2  FACID3	recycled to total acid phase leaving the ith separator (fraction of acid phase recycled)  ratio of acid phase leaving the ith separator in the organic stream forward to total acid phase leaving the ith separator (fraction of acid phase sent forward)  equivalent to FACID(1)  equivalent to FACID(2)

FAFR	fraction of total acid phase leaving separator 1 that appears in the external recycle from that separator
FAR	fraction of total acid phase leaving the separator that is recycled for the separator currently being calculated
FA1	equivalent to FA(1)
FA2	equivalent to FA (2)
FAS	equivalent to FA (3)
FA4	equi-valent to FA (4)
FA5	equivalent to FA (5)
FA6	equivalent to FA (6)
FD	extent of reaction for nitrosylsulfuric decomposition
FIN (I,J)	lb/hr of feed i to nitration vessel j
<b>FMAX</b>	maximum allowable volumetric flow of internal acid recycle
FMX	equivalent to FMAX
FØ (I)	ratio of organic phase leaving the ith separator with the organic stream forward to total organic phase leaving the ith separator (fraction organic phase forward)
FØRG (I)	ratio of organic phase leaving the ith separator with the soid recycle streams to total organic phase leaving the i <sup>th</sup> separator

equivalent to FØRG (1)

FØRG1

FØRG2	equivalent to FQRG(2)
FØRG3	equivalent to FØRG (3)
FØRG4	equivalent to FØRG (4)
FØRG5	equivalent to FØRG (5)
FØRG8	equivalent to FØRG (6)
FQ1	equivalent to FØ(1)
FQ2	equivalent to FØ(2)
FQ3	equivalent to FØ(3)
FQ4	equivalent to FG (4)
PQ5	equivalent to FØ (5)
roe	equivalent to FQ (6)
PP <b>Ģ</b>	fraction of organic phase leaving a separator that appears in the organic stream forward for the separator currently being calculated
FR (I)	ratio of external recycle stream to total recycle leaving the i <sup>th</sup> separator
FRAC	ratio of internal recycle stream to total recycle leaving the separator currently being calculated
FRAC1	equivalent to FRC (1)
FRAC2	equivalent to FRC (2)
FRACS	equivalent to FRC (3)
FRAC4	equivalent to FRC (4)
FRAC5	equivalent to FRC (5)

0	FRACE	equivalent to FRC (8)
	FRC (I)	ratio of internal recycle stream to total recycle stream leaving the ith separator
	FR1	equivalent to FR (1)
	FR2	equivalent to FR (2)
	FR3	equivalent to FR (3)
	FR4	equivalent to FR (4)
	FR5	equivalent to FR (5)
	FR6	equivalent to FR (6)
	PV (I)	volume of the ith nitrator (ou ft)
	FVKM	volume of the vessel currently being calculated
	FVKMS	volume of a separator (all separators have the same volume)
	FVKM1A	volume of nitrator 1A; equivalent to FV(1)
	FVKM1B	volume of nitrator 1B; equivalent to FV(2)
	FVKM2 FVKM3A	volume of nitrator 2; equivalent to FV (3) volume of nitrator 3A; equivalent to FV (4)
	FVKM3B	volume of nitrator 3B; equivalent to FV(5)
	FVKM4	volume of nitrator 4; equivalent to FV (8)
	PVKM5	volume of nitreter 5; equivalent to FV (7)
		<del>-</del>

FVKM6

G1 (I)

volume of nitrator 6; equivalent to FV (8)

ratio of Arrhenius coefficients in the kinetic rate expressions for reaction 1 and 2 in the ith nitrator

G10(I)	value of the Arrhenius coefficient in the kin stic rate expression for reaction 10 in the ith nitrator
G3 (I)	NOT USED
G38 (I)	value of the Arrhenius coefficient in the kinetic rate expression for reaction 3 in the i <sup>th</sup> nitrator under strong acid conditions
G48 (I)	NOT USED
G48 (I)	same as G38 (I) but for reaction 5
G5 (I)	value of Arrhenius coefficient in the kinetic rate expression for reaction 5 in the i <sup>th</sup> nitrator
G6 (I)	same as G5(I) but for reaction 6
Q7 (I)	same as G5(I) bur for reaction 7
G8 (I)	same as G5(I) bur for reaction 8
G9 (1)	NOT USED
I	DØ LØØP index
IC	counter for plotting intervals
ICALL	flag for first call to SUBROUTINE INPT
ICQUNT	equivalent to IC
IN	DG LGGP index for certain loops pertaining to nitrators
IPNCH	I/O device numbc. for punched cards
IPRNT	print control flag:
	0 summary printout only 1 debug printout of internal calculations during each integration step

IQUIT

number of stages being simulated

18

DØ LØØP index for certain loops pertaining

to separators

ISTGE

stage number

ITYPE

vessel type flag:

ITYPE=0 for nitrator ITYPE=1 for separator

IV

nitration vessel number

**IVSL** 

general vessel number

J

DØ LØØP counter and generally used integer

operator

**JACK** 

counter for integration time steps

K

DØ LØGP counter and generally used integer

operator

MW (I)

molecular weight of component i

NM

integer number of integration passes through the process (or iterations) required for a particular run. Also the integer number of integration time

steps required for the run

NN

integer number of iterations between summary

printouts

NØ (I)

notch setting on internal recycle gate valve for

separator i

NOTCH

notch setting for the separator currently being

calculated

NP

integer number of iterations between computations

of process performance indicators and other data

for plotting

N1	equivalent to NØ(1)
N2	equivalent to NØ(2)
N3	equivalent to NØ(3)
N4	equivalent to NØ(4)
N5	equivalent to NØ (5)
N6	equivalent to NQ(6)
P(I)	moles/hr of component i in the total organic phase leaving the vessel currently being calculated
PA	lb/hr of total acid phase leaving a separator for which acid phase specific gravity is being calculated
PF (I)	moles/hr component i in the organic phase of the organic stream leaving a separator
PI (I)	moles/hr component i in the total organic phase input to the vessel currently being calculated
PIR (I)	moles/hr of component i in the organic phase of the internal recycle leaving a separator
PNT (I,J)	noles/hr of component i in the total organic phase leaving nitrator j
PP	lb/hr of total organic phase leaving a separator for which organic phase specific gravity is being calculated
PS (I , J)	moles/hr of component i in the total organic phase leaving the j <sup>th</sup> separator
PV (I,J)	initial value of moles/hr component j in the total organic phas loaving vessel i

O	PXR (I)	moles/hr of component i in the organic phase of the external recycle leaving a separator
	P1A (I)	moles/hr of component i in the organic phase leaving nitrator 1A
	P1AI(I)	moles/hr of component i in the organic feed stream to nitrator 1A (moles/hr toluene to 1A)
	P1B (J)	moles/hr of component i in the organic phase leaving nitrator 1B
	P1BI(I)	moles/hr of component i in the organic feed stream to nitrator 1B (moles/hr toluene to 1B)
	P1S (I)	moles/hr component i in the total organic phase leaving separator 1
	P2 (I)	moles/hr component i in the organic phase leaving nitrator 2
	P2S (I)	moles/hr component i in the total organic phase leaving separator 2
	P3A (I)	moles/hr component i in the organic phase leaving nitrator 3A
	P3B (I)	moles/hr component i in the organic phase leaving nitrator 3B
	P3S (I)	moles/hr component i in the total organic phase leaving separator 3
	P4 (I)	moles/hr component i in the organic phase leaving nitrator 4
	P4S (I)	moles/hr component i in the total organic phase leaving separator 4
	P5 (I)	moles/hr component i in the organic phase leaving nitrator 5

P5S (I)	moles/hr component i in the total organic phase leaving separator 5
P6 (I)	moles/hr component i in the organic phase leaving nitrator 6
P68 (I)	moles/hr component i in the total organic phase leaving separator 6
Q	accumulation term used in calculation of nitrator heat loads
QA	volumetric flow of total acid phase leaving a separator for which acid phase specific gravity is being calculated
QN (I,J)	heat load in nitrator j calculated at the end of the i <sup>th</sup> plotting interval
QP	volument flow of total organic phase leaving a separator for which organic phase specific gravity is being calculated
QY (I) R (I)	fraction of nitric acid converted to nitronium ions in nitration vessel i NOT USED
PATES (I,J)	rate of reaction j in nitration vessel i
∂ <b>HØ</b> (I)	molar density of component i
нн01	equivalent to RHØ(1)
RHQ10	equivalent to PHØ (10)
RHØ11	equivalent to RHØ (11)
RHØ12	equivalent to RHQ (12)
RHQ13	equivalent to RHØ (13)
RHØ14	equivalent to RHØ (14)

RHQ2	equivalent to RHØ(2)
RHØ3	equivalent to RHQ (3)
RHQ4	equivalent to RHQ(4)
RHØ5	equivalent to RHØ5
RHØ6	equivalent to RHQ(6)
RHQ7	equivalent to RHQ (7)
RHØ8	equivalent to RHØ(8)
RHØ9	equivalent to RHQ(9)
RNS(I,J)	nitric to sulfuric acid ratio in nitration vessel j calculated at the end of the i <sup>th</sup> plotting interval (for acid phase only)
RNSS (I,J)	nitric to sulfuric acid ratio in separator j calculated at the end of the i <sup>th</sup> plotting interval (for acid phase only)
RP	coefficient on the molecular oxygen term in reaction 9
RR	ideal gas law constant
SA	adjustable constant. NOT USED
SB	equivalent to organic phase mass transfer coefficient (KMAP)
SC	equivalent to AKEQ4 in SUBROUTINE SUB
SD	NOT USED
SE	equivalent to AK9. NOT USED

equivalent to E9. NOT USED

SG

SH equivalent to AKEQ1 in SUBROUTINE SUB SI NOT USED equivalent to AKEQ3 in SUBROUTINE SUB SJ SPA accumulator term used in calculation of spent acid mass flow rate spent acid mass flow rate at the end of the ith SPENT (I) plotting interval (flow rate of acid phase of the external recycle stream leaving separator 1) SPO mass flow rate of dissolved organic in the spent acid accumulator term used in calculation of the mass SUM flow rate of crude TNT leaving separator 8 T temperature of nitration vessel currently being calculated TDNT mass flow rate of DNT in the total organic stream leaving separator 6 TCOX cumulative total moles/hr oxides of carbon (gas) leaving all vessels in the nitration section during a given iteration temperature of the ith nitration vessel TEMP (I) TI NOT USED TIME simulated elapsed time since beginning of run **TLBS** total lb/hr of acid phase leaving a separator on a nitrobody free basis. Used in computation of acid concentration indicators total moles/hr of stream i leaving a separator TM (I) currently being calculated

TNOX	total incles/hr oxides of nitrogen (gas) leaving all vessels in the nitration section during a given iteration
TR	reference temperature
TSEP(I)	computed temperature of the separator currently being calculated
TTCOX (I)	same as TCOX except that value is particular value at the end of the i <sup>th</sup> plotting interval
TTME	total time of process operation to be simulated
TTNOX (I)	same as TNOX except that value is particular value at the end of the i <sup>th</sup> plotting interval
TVL (I)	volumetric flow (cu ft/hr) of the ith stream leaving the separator currently being calculated
TW (I)	mass flow (lb/hr) of the $i^{th}$ stream leaving the separator currently being calculated
T1A	temperature of nitrator 1A (equivalent to TEMP(1))
TIB	temperature of nitrator 1B (equivalent to TEMP(2))
T2	temperature of nitrator 2 (equivalent to TEMP(3))
T3A	temperature of nitrator 3A (equivalent to TEMP(4))
ТЗВ	temperature of nitrator 3B (equivalent to TEMP (5))
<b>T4</b>	temperature of nitrator 4 (equivalent to TEMP(8))
<b>T</b> 5	temperature of nitrator 5 (equivalent to TEMP (7))
T6	temperature of nitrators (equivalent to TEMP(8))
VKM (I)	volume of the ith nitrator

では、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmのでは、100mmので

**VKMS** 

volume of a separator (same for all separators)

X (I)

elapsed simulated time at the end of the ith plotting interval (nomenclature is specific to plotting routine for plotting one dimensional variables as a function of time)

XX (I,J)

elapsed simulated time at the end of the i<sup>th</sup> plotting interval (this form of elapsed time is used for plotting variables of up to j dimension)

XYY(I)

moles/hr nitric acid in the acid phase converted to nitronium ions in nitration vessel i

## Subroutine INPT

The definitions of the following variables are identical to those in the main program: AF(I), AIR(I), AKMA, AKMATL, AK10, AK12, AK38, AK3W, AK4s, AK4W, AK5S, AK6S, AK7, AK8, AK9, ARRAY(I,J), AXR(I), A1S(I), A28 (I), A38 (I), A48 (I), A58 (I), A68 (I), CP (I), DH (I), DT, EAGP1, EAGP2, EAGP EAGP3, EAGP4, EAGP5, EAGP6, EGAP1, EGAP2, EGAP3, EGAP4, NGAP5. EGAP6, EX, E10, E21, E3S, E3W, E4S, E4W, E5S, E6S, E7, E6, E9, FAC1D1, FACID2, FACID3, FACID4, FACID5, FACID6, FA1, FA2, FA3, FA4, FA5, FA6, FIN(I,J), FMX, FORG1, FORG2, FORG3, FORG4, FORG5, FORG6. FQ1, FQ2, FQ3, FQ4, FQ5, FQ6, FRAC1, FRAC2, FRAC3, FRAC4, FRAC5, FRAC6, FR1, FR2, FR3, FR4, FR5, FR6, FVKM1A, FVKM1B, FVKM2, FVKM3A, FVKM3B, FVKM4, FVKM5, FVKM6, G1(I), G10(I), G3(I), G3S(I), G4(I), G48(I), G5(I), G6(I), G7(I), G8(I), G9(I), ICALL, IPRNT, JACK, MW(I), NM, NN, NP, N1, N2, N3, N4, N5, N6, PF(I), PIR(I), PXR(I), P1AI(I), P1BI(I), P1S(I), P2S(I), P3S(I), P4S(I), P5S(I), P6S(I), RATES (I.J), RHO1, RHO10, RHO11, RHO12, RHO13, RHO14, RHO2, RHO3. RHQ4, RHQ5, RHQ6, RHQ7, RHQ8, RHQ9, RP, RR, SA, SB, SC, SD, SE, SG, SH, SI, SJ, TCOX, TEMP(I), TNOX, TR, TSEP(I), TTME, T1A, T1B, T2, T3, T4, T5, T5,

## Remaining variables are defined as follows:

COMP (I,J) weight % component i in feedstock type j

DDL fractional part of a change in feed rate or nitrator temperature which is applied to the process during a given iteration

DL(I)

magnitude of the ith change in an indepenent variable

ICH (J)	integer code number for the independent variable type to which the j <sup>th</sup> change is being made
IKT	flag to indicate whether or not temperature changes have been made to the initial conditions
IPRNT	simulated time between summary printouts
IVS (J)	integer code number for the vessel to which the jth change in one of the independent variable types is made
NCHNG	number of given change which is to be made during the run
NOTCH (I)	notch setting on internal recycle gate value for the ith separator
NTME (I)	number of iterations between the beginning of a run the initiation of the ith change in an independent variable
NTVL (I)	number of iterations over which the magnitude of the ith change is to be equally distributed (used for ramping an independent variable)
TME	simulated time at which a given change is to be initiated
TVL	simulated time interval over which a given change is to be made
7.A	temperature coefficient in the exponent of the Arrhenius rate expression

#### Subroutine SUB

The definitions of the following variables are identical to those in the main program: A(I), AI(I), AKMA, AKMATL, AK10, AK12, AK38, AK3W, AK48, AK4W, AK58, AK68, AK7, AK8, CP(I), DH(I), DT, EX, E10, E21, E3W, E3S, E4W, E4S, E6S, E7, E6, FAII, FD, FMX, FPØ, E5S, FRAC, FVKM, G1(I), G10(I), G3(I), G3S(I), G4(I), G4S(I), G5(I), G3(I), G7(I), G8(I), G9(I), ICALL, IPRNT, ISTGE, ITYPE, IV, JACK, NM, NN, NOTCH, NP, P(I), PI(I), QY(I), RATES(I,J), RHØ1, RHØ11, RHØ12, RHØ13, RHØ14, RHØ2, RHØ3, RHØ4, RHØ5, RHØ6, RHØ7, RHØ6, RHØ9, RR, RP, SA, SB, SC, SD, SE, SG, SH, SI, SJ, T, TCOX, TI, TNOX, TR, TSEP(I), TTME, XYY(I)

The definitions of the following variables are identical to other variables defined for the main program:

Variable in SUB	Variable in main program
All thru All?	AI (1) thru AI (17)
A1 thru A17	A(1) thru A(17)
EAGP	EAP
EQAP	EØP
PI1 thru PI17	PI(1) thru PI(17)
P1 thru P17	P(1) thru P(17)

#### Remaining variables are defined as follows:

AINB	moles/hr nitrobody in the aggragate acid phase input to the vessel currently being calculated
AKA	general rate factor for all nitration reactions. Also if Bennett's strong acid rate expression is used, the coefficient of the besulfate ion concentration
AKB	coefficient of the sulfuric acid concentration in Bennett's rate expression
AKC	coefficient of the pyrosulfate ion concentration in Bennett's rate expression

AKEQ	equilibrium constant for the dissociation of sulfuric acid in an aqueous medium
AKEQOV	equilibrium constant for the dissociation of a mixture of sulfuric and pyrosulfuric acids to ionic species. NOT USED
AKEQ1	equilibrium constant for the dissociation of nitric acid in aqueous sulfuric acid
WKEG3	equivalent to AKEQ
AKEQ3	equilibrium constant for the dissociation of nitric acid in anhydrous sulfuric acid
AKEQ4	equilibrium constant for the recombination of nitronium and bisulfate ions in anhydrous acid mixtrues
AKMAP	organic phase mass transfer coefficient
AK1	computed value of AKEQ1 obtained from interval halving procedure
AK3	computed value of AKEQ3 obtained from interval halving procedure
AK4	computed value of AKEQ4 obtained from interval halving procedure
AL	moles/hr water in the total acid phase of the vessel current being calculated; can have negative value which then represents moles/hr SO <sub>3</sub>
AMNB	moles/hr nitrobody transferred from organic to acid phase by bulk mass transfer; negative value means transfer from acid to organic phase

same as AM1 but for mMNT

moles/hr aMNT transferred by bulk mass transfer

AM1

AM2

AM3	same as AM1 but for aDNT
AM4	same as AM1 but for mDNT
AM5	same as AM1 but for aTNT
AM6	same as AM1 but for .mTNT
aM7	same as AM1 but for TNP
AM8	same as AM1 but for TNBX
ANB	total moles/hr of nitrobody dissolved in the soid phase leaving the vessel currently being calculated
A804	combined moles/hr of SO <sub>3</sub> and H <sub>2</sub> SO <sub>4</sub> in the acid phase leaving the vessel currently being calculated
AT	total moles/hr of acid phase leaving the vessel currently being calculated
A11QA	concentration (moles/ft <sup>2</sup> ) of sulfuric acid in the acid phase of the vessel currently being calculated
В	initial concentration (moles/ft <sup>2</sup> ) of free sulfuric acid in the acid phase of the vessel currently being calculated
BC	3 + (3 x C)
C	initial concentration (moles/ft <sup>2</sup> ) of pyrosulfuric acid in the acid phase of the vessel currently being calculated
СВ	(.5 x B) + (1.5 x C)
CF	C-F
CHNO3	equilibrium concentration of nitric acid in the acid phase of the vessel currently being calculated

CHSC4	equilibrium concentration of bisulfate ions in the acid phase of the vessel currently being calculated
CH82O7	equilibrium concentration of pyrosulfate ions in the acid phase of the vessel currently being calculated
CH3O	same as CHNO3 but for water
CH28O4	equilibrium concentration of sulfuric acid in the acid phase of the vessel currently being calculated
CH3O	equilibrium concentration of hydronium ions in the acid phase of the vessel currently being calculated
CNO3	equilibrium concentration of nitronium ions in the acid phase of the vessel currently being calculated
cx	consolidation term for (ZA + YP)
CZ	consolidation term for (2B - YA + 1 + GX)
C5	moles/hr nitrosylfulfuric acid produced by oxidation reactions in the vessel currently being calculated
CP8	moles/hr nitrosylsulfuric acid which remain undecomposed in the vessel currently being calculated
מ	initial acid phase concentration of combined sulfuric acid plus pyrosulfuric acid in the vessel currently being calculated
DAL	rate of change of AL with respect to time in the vessel currently being calculated
DASQ4	rate of change of ASO4 with respect to time in the vessel currently being calculated
DA1	rate of change of A1 with respect to time in the vessel currently being calculated

DA12	rate of change of A12 with respect to time in the vessel currently being calculated
DA2	rate of change of A2 with respect to time in vessel currently being calculated
DA3	rate of change of A3 with respect to time in the vessel currently being calculated
DA4	same as DAS but for A4
DAS	same as DA3 but for A5
DA6	same as DA3 but or A8
DA7	same as DA3 but for A7
DA8	same as DA3 but for A8
DA9	same as DA3 but for A9
DRLTA	acid concentration $v \in \mathbf{m}$ in the weak acid nitration rate expression . NOT USED
DRLTA	
	rate expression. NOT USED rate of change of P1 with respect to time in the
DP1	rate expression. NOT USED rate of change of P1 with respect to time in the vessel currently being calculated
DP1	rate expression. NOT USED rate of change of P1 with respect to time in the vessel currently being calculated same as DP1 but for P16
DP1 DP10 DP2	rate expression. NOT USED  rate of change of P1 with respect to time in the vessel currently being calculated  same as DP1 but for P16  same as DP1 but for P2
DP1 DP10 DP2 DP3	rate expression. NOT USED  rate of change of P1 with respect to time in the vessel currently being calculated  same as DP1 but for P10  same as DP1 but for P2  same as DP1 but for P3
DP1 DP10 DP2 DP3 DP4	rate expression. NOT USED  rate of change of P1 with respect to time in the vessel currently being calculated  same as DP1 but for P10  same as DP1 but for P2  same as DP1 but for P3  same as DP1 but for P4
DP1 DP10 DP2 DP3 DP4 DP5	rate expression. NOT USED  rate of change of P1 with respect to time in the vessel currently being calculated  same as DP1 but for P10  same as DP1 but for P2  same as DP1 but for P3  same as DP1 but for P4  same as DP1 but for P5

DTA	acid phase adjusted integration time step
DTP	organic phase adjusted integration time step
D1	moles/hr of $\alpha$ -MNT that diffuse into the soid phase in the vessel currently being calculated
D10	moles/hr of toluene which diffuse from the bulk organic phase to the acid-organic interface
D2	same as D1 but for m-MNT
D3	same as D1 but for $\alpha$ -DNT
D4	same as D1 but for m-DNT
D5	same as D1 but for $\alpha$ -TNT
D6	same as D1 but for m-TNT
<b>D7</b>	same as D1 but for TNB
D8	same as D1 but for TNBX
ETA	overall mass transfer coefficient in the vessel currently being calculated
ETAA	acid phase mass transfer coefficient in the vessel currently being calculated
ETAP	organic phase mass transfer coefficient in the vessel currently being calculated
BTATL	mass transfer coefficient for toluene
F	initial acid phase concentration of nitric acid in the vessel currently being calculated
FA	volumetric fraction of aci ' phase in the vessel currently being calculated

FF (I)	cross sectional area fraction corresponding to the ith notch setting on the gate valve in the internal recycle line of the separator currently being calculated
FOV	constant set equal to one
PP	volumetric fraction of organic phase in the vessel currently being calculated
F10A	reaction rate factor for reaction 10 where $\alpha$ -TNT is the reactant for the vessel currently being calculated
F10M	same as F10A but for m-TNT
F3	reaction rate factor for reaction 3 in the vessel currently being calculated
F4	same as F3 but for reaction 4
F5	same as F3 but for reaction 5
F6	same as F3 but for reaction 6
F7A	reaction rate factor for reaction 7 in the vessel currently being calculated where $\alpha$ -DNT is the reactant
F7M	same as F7A but for m-DNT
F8A	reaction rate factor for reaction 8 in the vessel currently being calculated where $\alpha$ -DNT is the reactant
F8M	same as F8A but for m-DNT
F9A	Zero
F9M	Zero

GAMMA	acid concentration term used in all nitration rate expressions
KLM	flag to indicate which nitration region exists in the vessel being calculated
PNB	total flow (moles/hr) of organic phase nitrobody components leaving the vessel currently being calculated
PP	exponent of nitrobody-in-acid- solubility correlation
Q	fraction nitric acid converted to nitronium ions for the vessel currently being calculated
ŲA	volumetric flow of the acid phase leaving the vessel currently being calculated
QP	volumetric flow of the organic phase leaving the vessel currently being calculated
QT	total volumetric flow of combined internal and external recycle leaving the separator currently being calculated
RATE(I)	rate of the i <sup>th</sup> reaction in the vessel currently being calculated
R1	rate of reaction 1 in the vessel currently being calculated; equivalent to RATE(1) (moles/hr)
R10	same as R1 but for reaction 10; equivalent to RATE(10); also R10A + R10M
R10A	rate of reaction 10 in the vessel currently being calculated where $\alpha$ -TNT is the reactant
R10M	same as R10A but for m-TNT
R2	same as R1 but for reaction 2; equivalent to RATE(2)

R3	same as R1 but for reaction 3; equivalent to RATE(3)
R4	same as R1 but for reaction 4; equivalent to RATE(4)
R5	same as R1 but for reaction 5; equivalent to RATE(5)
R6	same as R1 but for reaction 6; equivalent to RATE(6)
R7	same as R1 but for the combined reaction 7; equivalent to RATE(7); also R7A + R7M
R7A	rate of reaction 7 in the vessel currently being calculated where $\alpha$ -DNT is the reactant
R7M	same as R7A but for m-DNT
R8	same as R1 but for the combined reaction 8; equivalent to RATE(8); also R8A + R8M
R8A	same as R7A but for reaction 8
R8M	same as R8A but for m-DNT
я9	same as R1 but for reaction 9; equivalent to RATE(9)
R9A	Zero
R9G	Zero
R9M	Zero
TKEL	temperature of the vessel currently being calculated (°K)
T1	A1 + P1
Т2	A2 + P2

Т3	A3 + P3
T4	A4 + P4
<b>T</b> 5	A5 + P5
<b>T</b> 6	A6 + P6
T7	A7 + P7
T8	A8 + P8
w	initial acid phase concentration of water in the vessel currently being calculated (moles/ $\hat{\pi}^s$ )
x	consolidation term used in the integration sequence of organic species in the acid phase of the vessel currently being calculated
XEQA	equilibrium solubility of nitrobody in the acid phase of the vessel currently being calculated (moles/hr)
ХL	lower limit of an ionic species concentration which is applied during internal halving procedure
χυ	upper limit of an ionic species concentration which is applied during internal halving procedure
XUP	upper limit of HS <sub>2</sub> O <sub>7</sub> concentration (resulting from dissociation according to equation 19) which is applied during internal halving procedure
X1	mole fraction $\alpha$ -MNT in the acid phase of the vessel currently being calculated
X13	mole fraction water in acid phase of the vessel currently being calculated
X14	mole fraction sulfur trioxide in the acid phase of the vessel currently being calculated

X2	mole fraction m-MNT in the acid phase of the vessel currently being calculated
<b>X5</b>	mole fraction $\alpha$ -TNT in the acid phase of the vessel currently being calculated
X6	mole fraction m-TNT in the acid phase of the vessel currently being calculated
жэ	mole fraction nitric acid in the acid phase of the vessel currently being calculated
Y	consolidation term used in the integration sequence for organic species in the soid phase of the vessel currently being calculated; also used for initial concentration (moles/ft <sup>3</sup> ) of free sulfuric in the soid phase of the vessel currently being calculated
YA	ratio of AMNB to ANB
YP	ratio of AMNB to PNB
Z	initial acid phase concentration of nitric acid which is used under strong acid nitrating conditions for which water is present; also used as a consolidation term in the integration sequence for organic species in the acid phase
ZA	enthalpy of the contents of a separator (BTU/hr - °C)
<b>2B</b>	rate of heat generated in a separator due to reaction (BTU/Hr)
ZD	A9-A17: undissociated acid phase nitric acid remaining at equilibrium in the vessel currently being calculated (moles/hr)

## Subroutine SUB2

The definitions of the following variables are identical to those in the main program: AKMA, AKMATL, AK10, AK12, AK3S, AK3W, AK4S, AK4W, AK5S, AK6S, AK7, AK6, CP(I), DH(I), E10, E21, E3S, E3W, E4S, E4W, E5S, E6S, E7, E8, FD, DMX, QA, QP, RHO(I), RP, RR, SA, SB, SC, SD, SE, SG, SH, SI, SJ, TR

## Remaining variables are defined as follows:

AQ(I)	moles/hr component i in the total acid phase leaving the separator for which recycle para- meters are currently being calculated
AT	total moles/hr of acid phase leaving the separator for which recycle parameters are currently being calculated
EAØP	molar ratio of acid to organic phase in the organic stream leaving the separator for which recycle parameters are currently being calculated
EÇAP	molar ratio of organic to acid phase in either of the recycle streams leaving the separator for which recycle parameters are currently being calculated
FAR	fraction of acid phase that is recycled in the separator for which recycle parameters are currently being calculated
FAR1	fraction of acid phase sent forward from the separator for which recycle parameters are currently being calculated
FF (I)	identical to definitions given in subroutine SUB
Frø	fraction of organic phase sent forward from the separator for which recycle parameters are currently being calculated

FPG1 fraction of organic phase which is recycled in

the separator for which recycle parameters are

currently being calculated

FRAC ratio of internal recycle stream to total recycle

streams leaving the separator for which recycle

parameters are currently being calculated

NOTCH notch setting on the internal recycle gate valve

for the separator for which recycle parameters

are being calculated

PØ moles/hr component i the total organic phase

leaving the separator for which recycle para-

meters are currently being calculated

PT moles/hr total organic phase leaving the separator

for which recycle parameters are currently being

calculated

QT volumetric flow (ft<sup>8</sup>/hr) of combined recycle

streams leaving the separator for which recycle

parameters are currently being calculated

### Subroutine SUB1

The definitions of the following variables are identical to those in the main program: AI(J), AKMA, AKMATL, AK10, AK12, AK38, AK3W, AK48, AK4W, AK5S, AK6S, AK7, AK8, AV(I,J), CHAMXØ, CP(I), DFAMXØ, DH(I), DT, EX, E10, E21, E38, E3W, E48, E4W, E58, E68, E7, E8, FD, FMX, G1(I), G10(I), G3(I), G3S(I), G4(I), G4S(I), G5(I), G6(I), G7(I), G8(I), G9(I), ICALL, IPRNT, IVSL, JACK, NM, NN, NP, PI(I), PV(I,J), QY(I), RATES(I,J), RHØ1, RHØ10, RHØ11, RHØ12, RHØ13, RHØ14, RHØ2, RHØ3, RHØ4, RHØ5, RHØ6, RHØ7, RHØ8, RHØ9, RP, RR, SA, SE, SC, SD, SE, SG, SH, SI, SJ, TCOX, TNOX, TR, TSEP(I), TTME, XYY(I)

### Remaining variables are defined as follows:

AØ (I)

moles/hr component i in the acid phase of the vessel for which component % changes are currently being calculated

DFA (I) acid phase component i % change during NN

iterations in the vessel currently being calculated

DFAMX maximum % change in an acid phase component

in the vessel currently being calculated

DFP (I) same as DFA (I) but for organic phase

DFPMX same as DFAMX but for organic phase

PØ(I) moles/hr component i in the organic phase of

the vessel for which component & changes are

currently being calculated

R1-R10 NOT USED

### Function ENTH

The definitions of the following variables are identical to those in the main program: AKMA, AKMATL, AK10, AK3S, AK3W, AK4S, AK4W, AK5S, AK6S, AK7, AK8, CP(I), DH(I), E10, E12, E3S, E3W, E4S, F4W, E5S, E6S, E7, E8, FD, FMAX, RHQ1, RHQ10, RHQ11, RHQ12, RHQ13, RHQ14, RHQ2, RHQ3, RHQ4, RHQ5, RHQ6, RHQ7, RHQ8, RHQ9, RR, SA, SB, SC, SD, SE, SG, SH, SI, SJ, TR

#### Remaining variables are defined as follows:

A(I) moles/hr component i in the acid phase of the

stream for which the enthalpy is currently

being calculated

AX (I) moles/hr component i in the total acid phase

leaving a vessel for which the enthalpy of a particular leaving stream is currently being

calculated

CP11 equivalent to CP (11)

CP12 equivalent to CP (12)

CP13 equivalent to CP(13)

CP14 equivalent to CP (14)

DLT	temperature of the vessel (relative to 20°C) for which the enthalpy of a particular exiting stream is currently being calculated
ENTH	enthalpy of the stream currently being calculated (kcal/hr)
enth <b>ø</b>	enthalpy of mixing of the acid components in the acid phase of the stream currently being calculated based on the zero nitric curve of the McKinley and Brown data (kcal/lb)
ENTH2	enthalpy of mixing of the acid components in the acid phase of the stream currently being calculated based on the 20% nitric curve of the McKinley and Brown data (kcal/lb)
F	adjusted weight fraction of nitric soid plus sulfuric acid in the acid phase of the stream currently being calculated on a nitrobody free basis
LBS	lb/hr of nitric acid, sulfuric acid, water (or sulfur trioxide) in the acid phase of the stream currently being calculated
LBSE	lb/hr of nitric acid and equivalent sulfuric acid in the acid phase of the stream currently being calculated
P(1)	moles/hr component i in the organic phase of the stream for which the enthalpy is currently being calculated
PX (I)	moles/hr component i in the total organic phase leaving a vessel for which the enthalpy of a particular leaving stream is currently being calculated
т	moles/hr combined organic and acid phase nitro- body component in the stream for which enthalpy is currently being calculated

TP	temperature of the vessel for which the enthalpy of an exiting stream is being calculated (°C)
WTA	LBSE/LBS; nitrobody free weight fraction of equivalent sulfuric plus nitric acid in the acid phase of the stream for which the enthalpy is currently being calculated
WTN	weight fraction of nitric acid in the equivalent sulfuric plus nitric acid of the acid phase of the stream for which the enthalpy is currently being calculated
X (I)	the ith data point on the zero % nitric curve from the enthalpy concentration data of McKinley and Brown
XA1	FACID, FA or 1 dependent on the origination point of the stream for which the enthalpy is currently being calculated
XA2	FR, FRAC or 1 dependent on the origination point of the stream for which the enthalpy is currently being calculated
XP1	FØ, FØRG or 1 dependent on the origination point of the stream for which the enthalpy is currently being calculated
XP2	FR, FRAC or 1 dependent on the origination point of the stream for which the enthalpy is currently being calculated
Y(I)	the ith data point on the 20% nitric curve from the enthalpy concentration data of McKinley and Brown

## APPENDIX E

Dynamic Simulation--input Data Format

Card	Data Read		Format
1	AK12, AK3W, AK4W, AK88, AK65, AK7	, AKB, AKLO	8F10.0
2	AK38, AK40, E38, E48		8 <b>F1</b> 0.0
3	E21, M3W, E4W, E58, E68, E7, E8,	<b>E1</b> 0	8 <b>F10.0</b>
4	9A, 5B, 9C, 8D, 6E, 9G, SH, 9I		8 <b>F</b> 10.0
5	15, RR, TR, AKMA, AKMATL, FD, RP		8 <b>r</b> 10.0
6, 7	DH (I), T=1, 10		8 <b>F10.0</b>
8, 2	RHM (1), 140, 10		8F10.0
10, 11	CP (I), I-1, 14		8710.0
12 - 14	ALA (I), I=1, 17	5m16.7/5m16.7/4m16.7.	3 <b>F</b> 1.0.0
14 - 15	PlA (1), Jul., 17	5m10.7/5m16.7/4m16.7,	3 <b>F</b> 10.0
17 - 1)	AlB (I), I=1, 17	5m14.7/5m14.7/4m14.7,	3T10.0
50 - 95	P1B (1), 1-1, 17	5m16.7/5m16.7/hm16.7,	3 <b>F10.</b> 0
23 - 25	Als (I), I=1, 17	5E16.7/5E16.7/4E16.7,	3 <b>F1</b> 0.0
2" - 28	P18 (1), Tw1, 17	5m16.7/5m16.7/4m16.7,	3 <b>F10.</b> 0
29 - 31	A2 (7), I+1, 17	5m16.7/5m16.7/4m16.7,	<b>3F10.</b> 0
32 - 3h	P2 (T), T-1, 17	5E16.7/5E16.7/ E16.7,	3 <b>71</b> 0.0
35 - 37	A23 (1), I=1, 17	5E16.7/5E16.7/4m16.7,	3730.0
38 - 40	F21 (1), I=1, 17	5m16.7/5m16.7/4m16.7.	3 <b>F</b> 10.0
41 - 43	*3% (I), T=1, 17	5m16.7/5m16.7/4m16.7.	3 <b>F</b> 10.0
44 - 46	P3A 'I), I=1, 17	5E16.7/5E16.7/4E16.7.	3 <b>F1</b> 0.0
47 - 49	A3D (I), I=1, 17	5m16.7/5m16.7/4m16.7,	3710.0
50 - 52	P3B (I), I=1, 17	5m16.7/5m16.7/4m16.7	3710.0
53 - 55	A39 (I), I=1, 17	5816.7/5816.7/4816.7	3710.0

Ü	Card	Data Read	Format	
	56 <b>- 58</b>	P3S (I), I=1, 17	5E16.7/5E16.7/4E16.7, 3F10.0	
	59 - 61	A4 (I), I=1, 17	5m16.7/5m16.7/4m16.7, 3m10.0	
	F2 - 54	Ph (I), <b>1-1</b> , 17	5m16.7/5m16.7/4m16.7, 3m10.0	
	65 - 67	A48 (I), I=1, 17	5e16.7/5e16.7/4e16.7, 3f10.0	
	69 - 70	Phs (I), I-1, 17	5e16.7/5e16.7/4e16.7, 3F10.0	
	73, - 73	A5 (I), I=1, 17	5E16.7/5E16.7/4E16.7, 3F10.0	
	71 - 75	' P5 (I), I=1, 17	5E16.7/5E16.7/4E16.7, 3F10.0	
	77 - 79	A58 (I), I=1, 17	5e16.7/5e16.7/4e16.7, 3f10.0	
	85 + 82	P5S (I), I=1, 17	5E16.7/5E16.7/4E16.7, 3F10.0	
	93 - 85	A6 (I), I=1, 17	5E16.7/5E16.7/4E16.7, 3F10.0	
	94 <b>-</b> 88	P6 (I), I=1, 17	5m16.7/5m16.7/4m16.7, 3m10.0	
	<i>የ</i> ኃ • ባኒ	A69 (I), I≈1, 17	5m16.7/5m16.7/4m16.7, 3m10.0	
	72 - 54	PGS (I), I=1, 17	5916.7/5E16.7/4E16.7. 3F10.0	
	<b>15</b>	N1, N2, N3, N1, N5, 116	611	
	94	enø (I), I=1,6	8F10.0	
	77	<b>π6</b> Λ (I), I=1, 6	8F10.0	
	98	VKM (I), I=1, 8	8F10.1	
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	100	TEMP (I), I=1, 8	8 <b>r</b> 10.0	
	101	тх	8 <b>F10.</b> 0	
	102 - 109	(FIN(I,J), J= 1, 8), I=	1, 7 SF10.0	
	1.09 - 118	(CØMP(J,I), I=1, 14), J	=1, 5 8F10.0/6F10.0	
	11?	IPRNT, TIME, DT, DPRNT	15, 3F10.0	
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# APPENDIX F

Steady State Simulation--Program Listing and Output

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PROGRAM TWEST

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V= V+   V= V+   D01444 = v 6  D01444 = v 6  D01444 = v 6  D111=20V  v 10    CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v 6  CAL   = 0V  v	TMTSS	2
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SALFURIC - OLEUM SYSTEM OREGION 13     PHTSSS       If LAM, GT., Soal (4)     GO TO 2737     PHTSSS       Condons     PHTSSS       Condons     PHTSSS       Condons     PHTSSS       Condons     PHTSSS       Condons     PHTSSS       Condons     PHTSSS       Condons     PHTSSS       Condons     PHTSSS       Condons     PHTSSS       Condons     PHTSSS       Condons     PHTSSS       Condons     PHTSSS       Condons     PHTSSS       SALCAMIC - OLEUM SYSTEM OREGION 33     PHTSSS       PHTSSS     PHTSSS       Reference     PHTSSS       PHTSSS     PHTSSS       Reference     PHTSSS       Reference     PHTSSS       PHTSSS     PHTSSS       Reference     PHTSSS       PHTSSS     PHTSSS       Reference     PHTSSS       PHTSSS     PHTSSS       PHTSSS     PHTSSS       PHTSSS     PHTSSS       PHTSSS     PHTSSS       PHTSSS     PHTSSS       PHTSSS     PHTSSS       PHTSSS     PHTSSS       PHTSSS     PHTSSS       PHTSSS     PHTSSS       PHTSSS     PHTSSS </td <td>u</td> <td></td> <td>INTSSS</td> <td>~</td>	u		INTSSS	~
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[0012],735,50,236630R [132],375,50,236630R [100,225,200,000]	}63-4718-81512-991_8-8-254674 <b>939</b> 85-1-84A474878-8-75339487[-4-8 <b>44E-4</b> +7L1-	S S S S S S S S S S S S S S S S S S S	2225
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Fluef 19 GOTOL CONTIME FAALICED SIDER/(TRID-TAIR)		1455 14155 14155 14155	1655 1655 1655 1655
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Secure distribution

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F(FA2.6T.) . FAB=).  FAQ1=1FAQ  FPA=1FAQ=(SEP! -AI/PT  FPA=1FAQ=0.0.0.0.0.0.0.0.0.0.0.0.0.0.0.0.0.0.0.	FPD:1.  F(F=0.ft.l.) FPC=1.  F(F=0.ft.l.) FPC=1.  F(F=0.ft.l.) FPC  FPC  FPC  FPC  FPC  FPC  FPC  FPC	f pic_sf uil seff (100TC+) / OT If (Frid_c, 6T = 1 - ) Frid_c=1 - If (Frid_c, 6T = 1 - ) Frid_c=1 - If (Frid_c, 7T = 1 - ) Frid_c, EACP (15EP) - PT + AT + GT + F (100TCH) + QA + QP +	f FPAX. f FPAX.FBA.FBA.FBA.FPO.FPOI f FPAX.FC EQAP.EAGP.PT.AT.GT.FF.BA.GP.FMAX**FT10.4/ c * FPAC.FR.FAR.FAR.*FPO.FPOI**6F10.5) A POI.FET.B.14		CONTINUE fig10/045 DOI-4-21-10 RAFS(IN-J)=B(J) CONTINUE		FORWATE 17**MITSATION VFSSEL**IZ**  C Sx 12**IESATIONS 2**13)  GOTOIS** IF (15E2**E.5.) CO TO 15:0  IF (15E2**E.5.) EO TO 15:0  FORWATE (2.15**15E**E**E**E**E**E**)*ICOUNT  FORWATE (3.15**15E**E**E**E**E**E**E**E**E**E**E**E**E**	2443,7x 2444,7x 2445,7x 2446, 444506,6x 34412,6x 2441,7x 2 7x 2444,7x 245,7x 2465,7x 4x 64001,33, 4x 54001,5,5x	DOISGIELSTA AVICTURESCI PVITTURESCI THE FOLLGHING CALCULATIONS ARE TO CHECK THE AGREEMENT WITH THE CORIGINAL EQUATIONS WITHTER CHECK (B) 60 TO 1234 SPWARFOLLD
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7-5-7-5-5	**************************************	THISS	1524
-0~:0		TNTSS	1528
, ~ . ~	77=+1-6-79-4018/3-14-8-14-1-6-88-4113/41-1-1-18-14-14-14-14-14-1-1-1-1-1-1-1-1	TNTSS	15%
	do++*6	18755	1531
י בי ט		TRISS	15 25
u <sup>_</sup> u	WPITE IS. 7001 IV. TKEL. AT. SAMP. SAMPC. PINB. PT. A.P. (AI(II. IFI. 141.	THISS	1534
·	[ [P][]]+[=]+]+]+]+ 	18755 THTSS	15.35
	Fla.4.51.05440Co.Fl4.4/* pp@o.Fl4.4.51.0Fl0.Fl4.4/* A 0.14F9.2/	TNTSS	1537
	e p e.icf4.2/a Alo.icf4.2/e Plt.16f9.2}	1.1755	223
AME -CAME		TWISS	3,5
Cipres.	ì	THISS	3
) 311es	##11E16.7561 #1.#2.#3.#4.#5.#6-#7.#8.#10.#96110.#96.#PeC5-C5F-A566.	TMTSS	1542
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_		TWISS	2
751 00(1)=6.		Turs	
6010352		THTSS	35
350 EIA=1./	ETA=1./(1./ETAP.1./(ETAA*AEQA))	TMTSS	3.51
ZA=£TA/PNS		TKTSS	
78=E11/AMB		TWTSS	1553
22(1)00	0(1)=240P(1)-280A(1)	<b>14155</b>	155
5-405 682		TMTSS	555
Table Car		THISS	1557
	F (Appl) 37 - 331 - 331	THISS	1558
136 Casamph/aug	A/Aug	TMTSS	1559
8	;	TWISS	
251 01 02		TATES	3
		THISS	1563
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_	117 [=].8	SSIMI	5
487=(})#A FEF	AP(() = (APA()) • (APA())	THISS	1567
411=41=41 411=41=41	(1) -636-411 -00(1) -44(1)	TKTSS	156
901=014-22		TWISS	3
(F) Your SELEC	Total Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control Control	14155	
MD 1 E (4.3)	MOTTE(4.382)	TWTSS	1572
TT=LIT	117=11(7) -(-1)-4(1) -(-2)-4(1) -(-2)-4(1) -(-2)-4(1) -(-2)-4(1) -(-2)-4(1)	TMTSS	1573
AXAEAT	11421 [4] - C. L. O. L. (2) - F. 6-1 [4] - 67 (2 L. (4) - 620 - A. (4) - 630 [4] - A. (4)	TKTSS	1574
AKS=AT	ars=al(s)+csc=a())-4,-c,pe=(s)+00(s)+am(5)	TATES	5757
18=948		TWT55	1577
ALASAT	118 11 (4) -1, -6, 00 (4) (5) -4 (6) -00 (8) -44 (P)	STITES.	157

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SUBPOUTINE VESSFL

			LHISS	
	6	0013=01000 (4.0)	TWTSS	1
678	a	000=(11,+007+14,5+008+0,+0016)*FD	TWTSS	1582
	d	PP9C=G0G*(A(3) +A(4))	14155	1543
	200	002201=1.8	TNISS	1584
	220 PX	PX(I)=DI(I)-DQ(I)-W(I)	THTSS	1545
	_	PX(1)=PX(1)+018-R2	THISS	1586
635	)Xd	PX(2)=PX(2)+R2	THTSS	1587
	C X C	PX3=1,51*4(11)*4(9)*PT/(AT*4(11))	TMTSS	1586
	PX 6	3X9=A1(9)+P1(9)-P(9)-D1F-G3D*A(1)-G4D*A(2)-G5G*A(3)-66G*A(4)-	TNTSS	1589
	1	[ ]].eco7-]?.5+AAA-9.+Ac]&-12.5+D096+(.33+.67*RP)+RR9	INTSS	1596
	pxi	610-(01) -013	THISS	1591
949	AXI	AXI2=AI(12)+(1,-F0)+(1, -R07+14.5+PRE+9.+R910)	THIES	1592
•	AKL	AKL=A1(13)-A1(14)+010+630-A(1)+640-A(2)+656-A(3)+666-A(4)+	TWTSS	1593
	) I	C 15.+007+14.5+008-(.67.33-00) -009+1200010	THTSS	15%
	da.	HPITE (4.155) 10.010 xEQ4.4H.AMMP.AIMB.AXI.AX2.AX3.AX4.AX5.AX6.	THISS	1595
	3	C AFT-AKH-AKH-AKI-ON-PK9-PKIS	TNTSS	15%
645	355 FOR	FORMATI'S DOSDIGS SELECT SASTED STORES AND SELO 43X PARKES		1597
	<u> </u>	C FIR.4. TX. +21NR+.FIB.4/* AKI-9+.9FIB.4+* AXI2.4XL+.2FIB.4/		1596
	٤	(+ px]-10+16F10*4)	TMTSS	1599
	\$	WPITE(6.751) GI-G3.60.07.66.67.68.69.618.630.640.656.666.	THISS	1606
	3	C 676,6AP,696,610F	THTSS	1691
959	761 FOR	FORMAI(* 61.63.64.65.64.67.66.67.68.69.9149/1199/12.2/	TMTSS	1602
	·	C * G10.G40.656.666.666.676.G8P.696.61@P*/1X8F12.4)	THTSS	1603
	1234 AFT	noil 38	TNTSS	1664
	•	IF (110) 162,162,174	THISS	1605
	176 001	001751=1.22	THTSS	1686
655		V(I)=Vn(I)+FACT*(V(I)-VD(I)	TNTSS	1607
		50Tn162	THTSS	1600
	154 179	TR= TR+	TKTSS	1609
	FAC	FACT#9.59F1CT	TNTSS	1610
	141	IF (ITR-5) LA.10.1690	TMTSS	1611
999	9	#PITE(6.167) IV	TNTSS	1612
,	FO4	FORMATIC THONESSEL-13- 19H FAILED TO CONVERGE)	IMTS.	1613
	100	9169	THISS	1614
	Í	5		717

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FTR 4.1-PSR367				AMMAY									•												ARRAY	MERLY															AMMA			
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				*		1113	A113	AIIS	A117	AI3	115	A 19	AKEO	AKE 03	AKE 03		AKIO	PK3	N. C.	7	1	z		l	AK	4110	21xv	AXS	AX7	A16	A13	714 124	, ন্	¥3	2 }	£ 2	5	ь	100	8 8	8	ğ	85	ij
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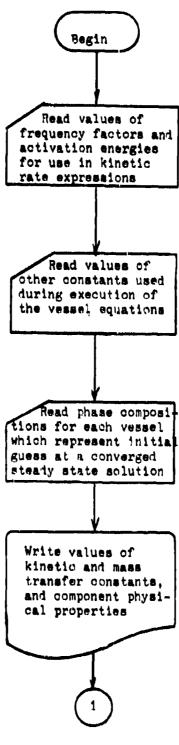
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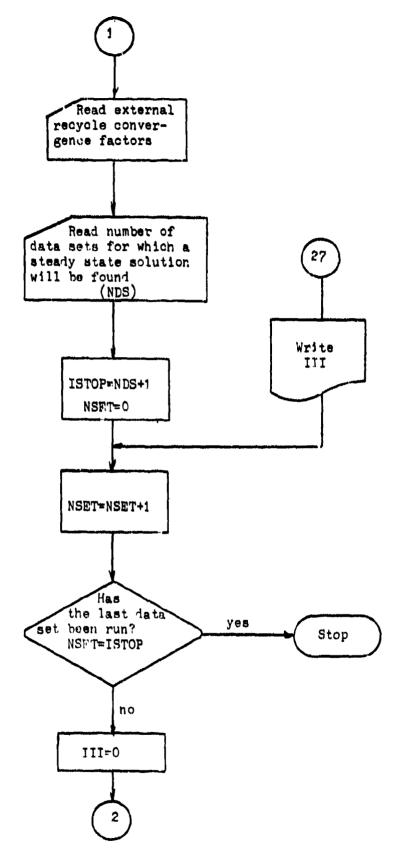
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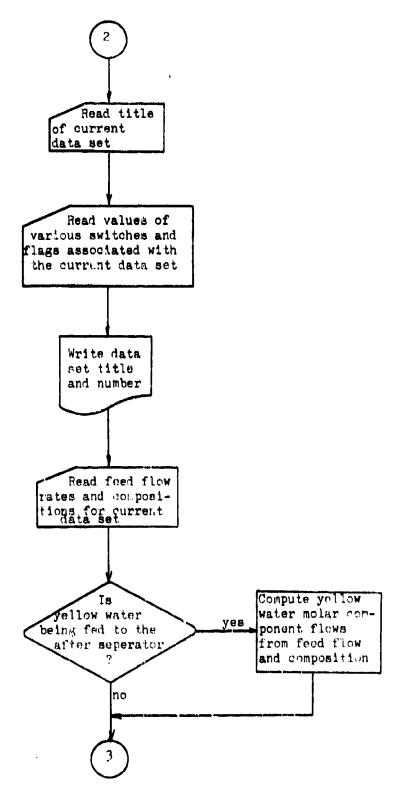
## APPENDIX G Steady State Simulation--Program Flowchart

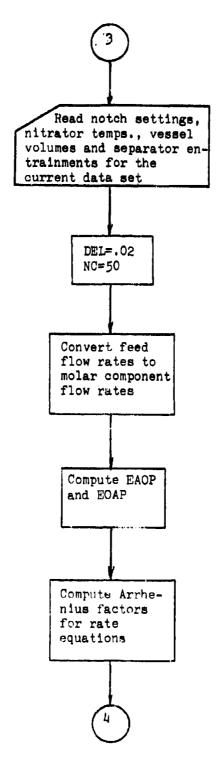
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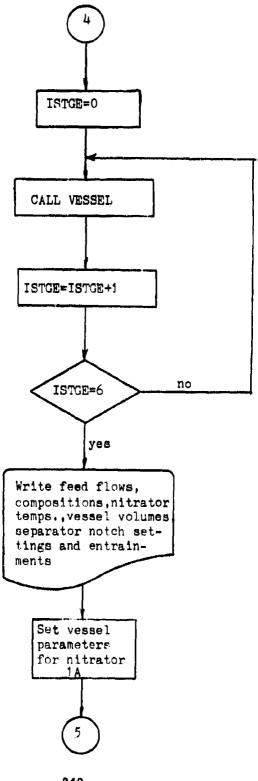


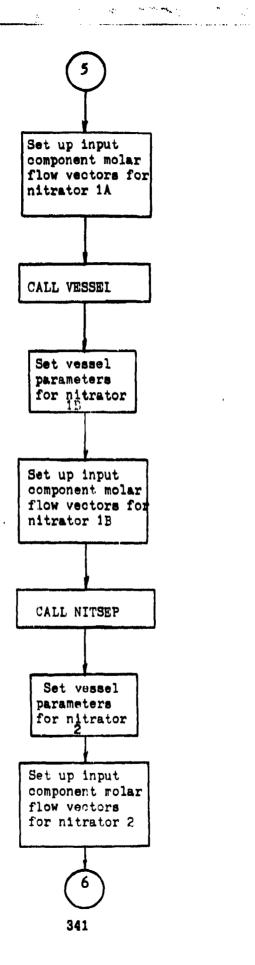


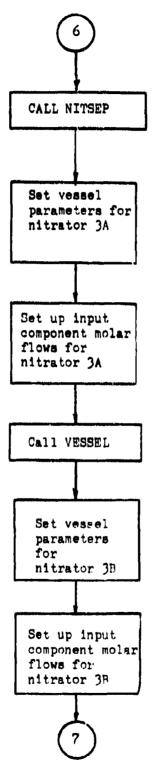
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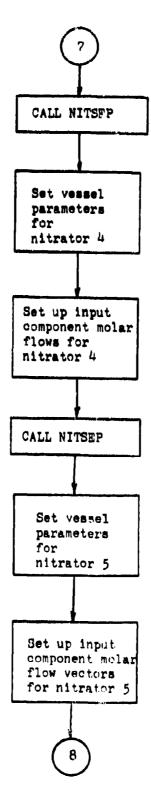


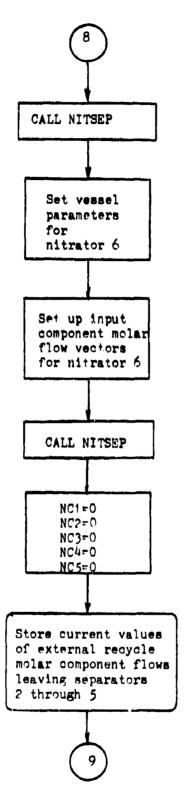


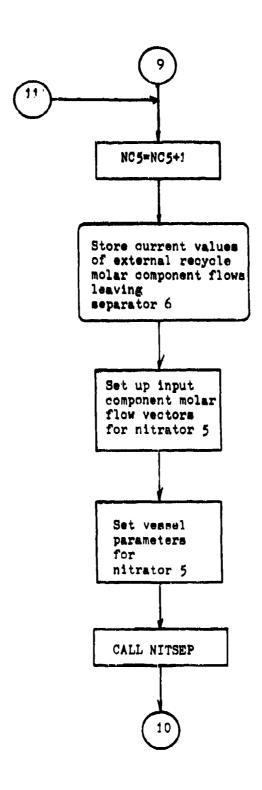


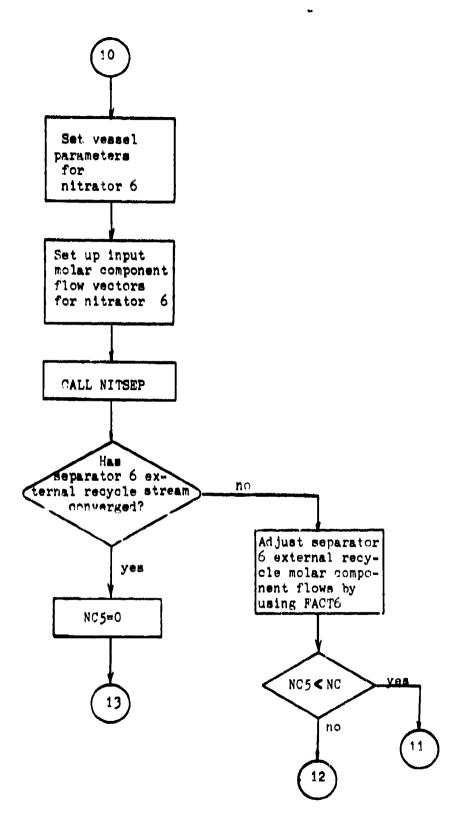






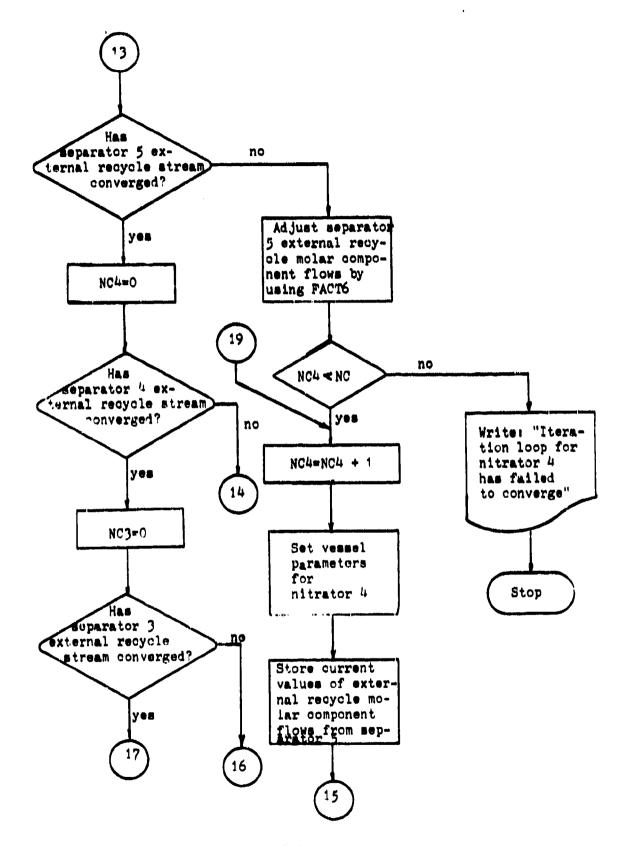


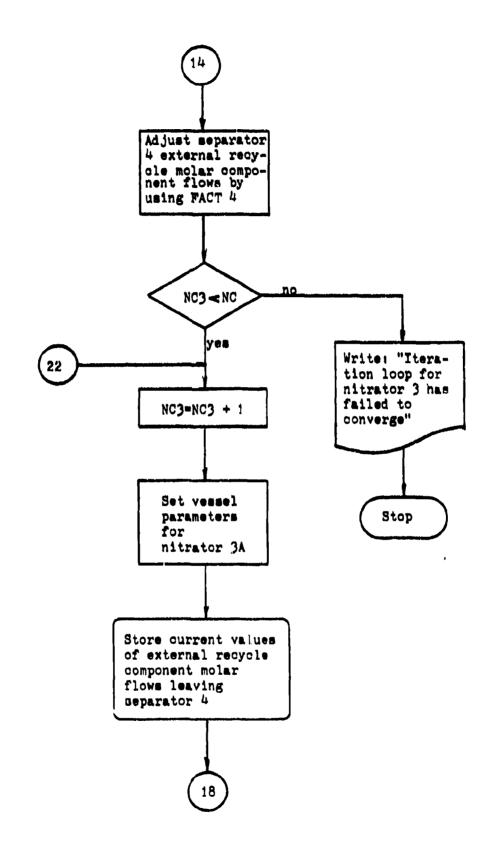


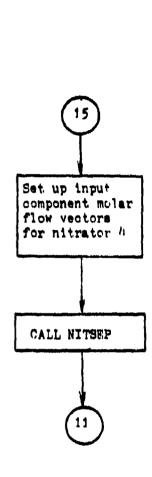


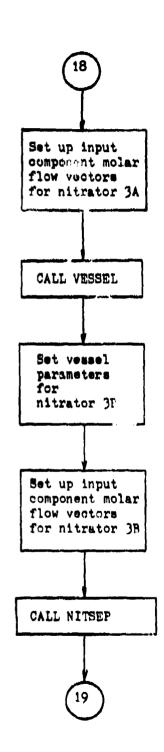


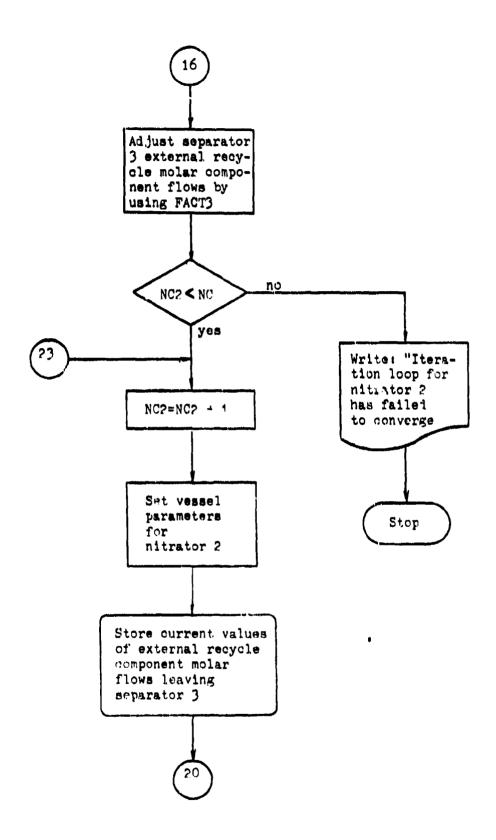
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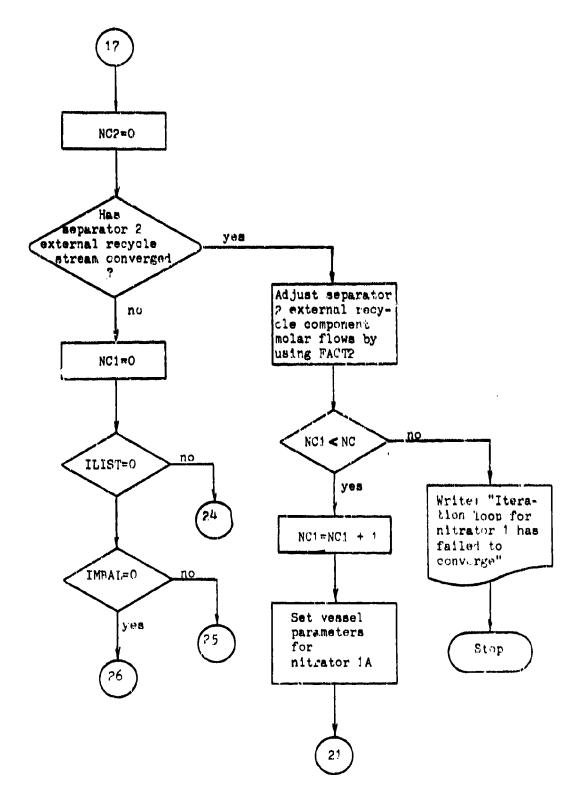


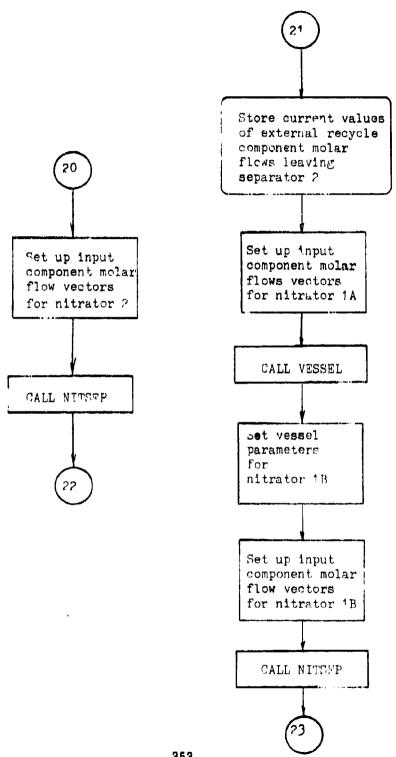


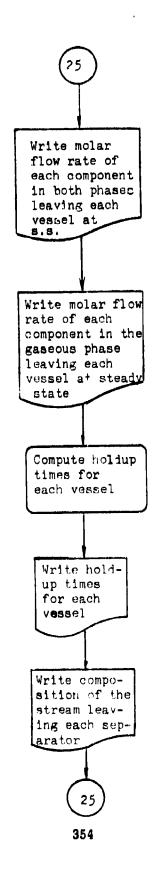


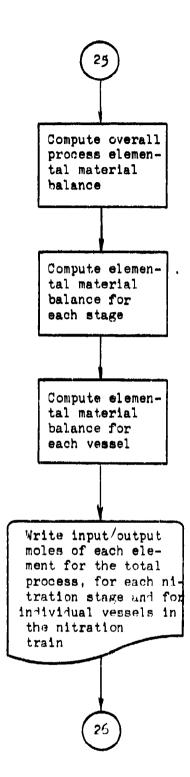


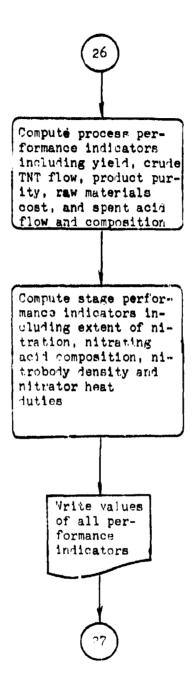
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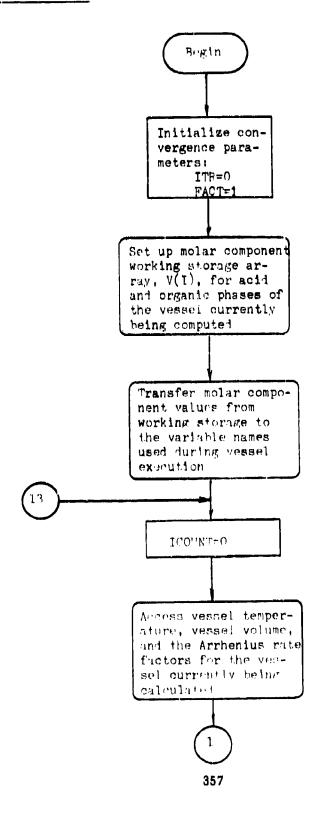


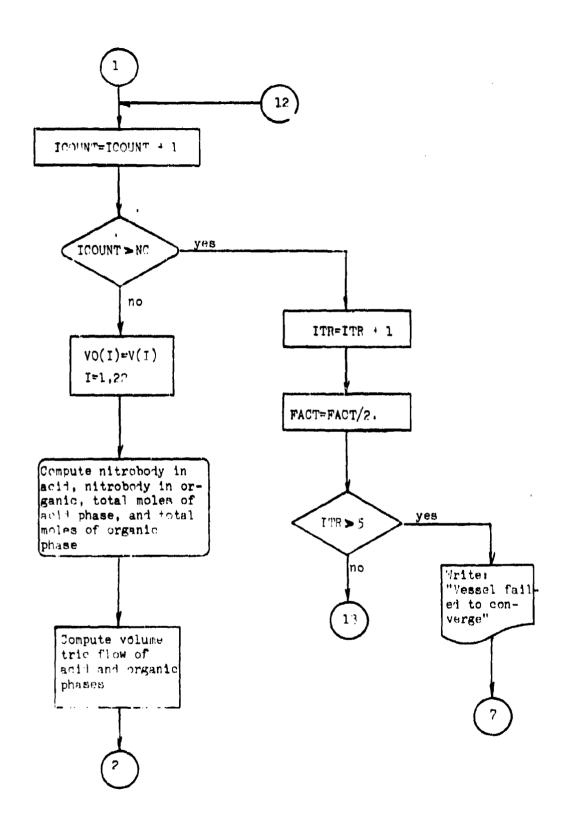


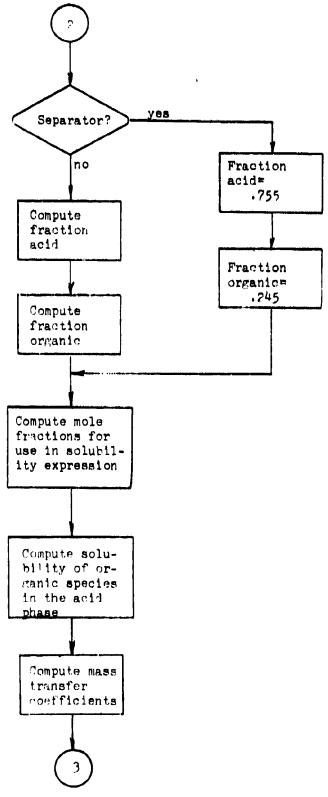


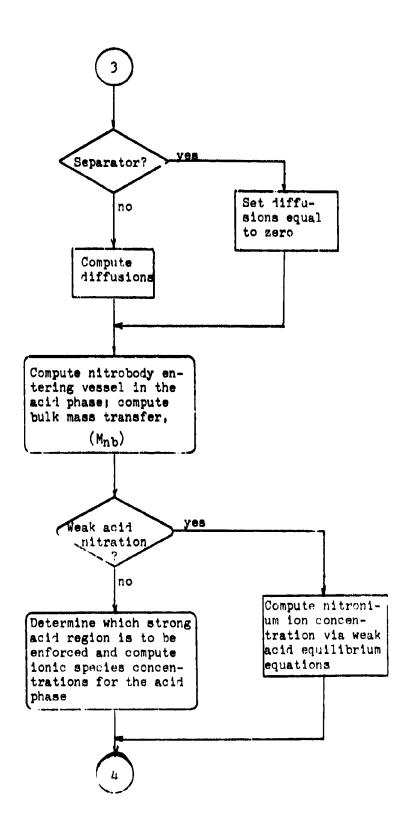


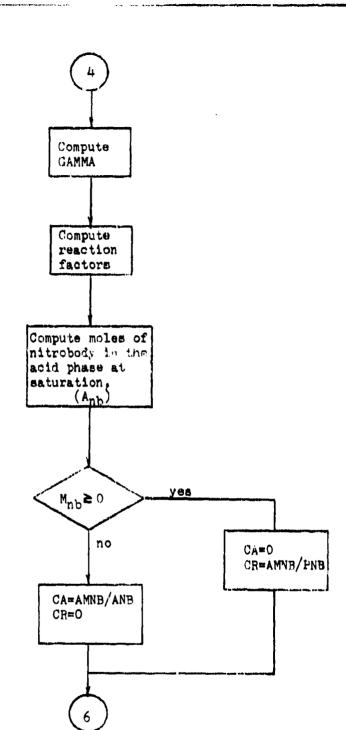




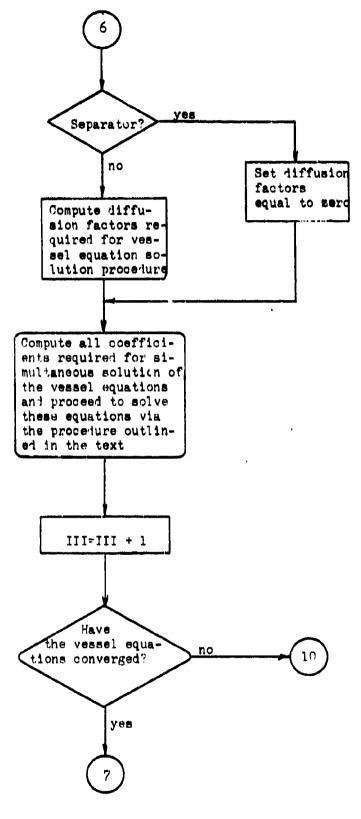


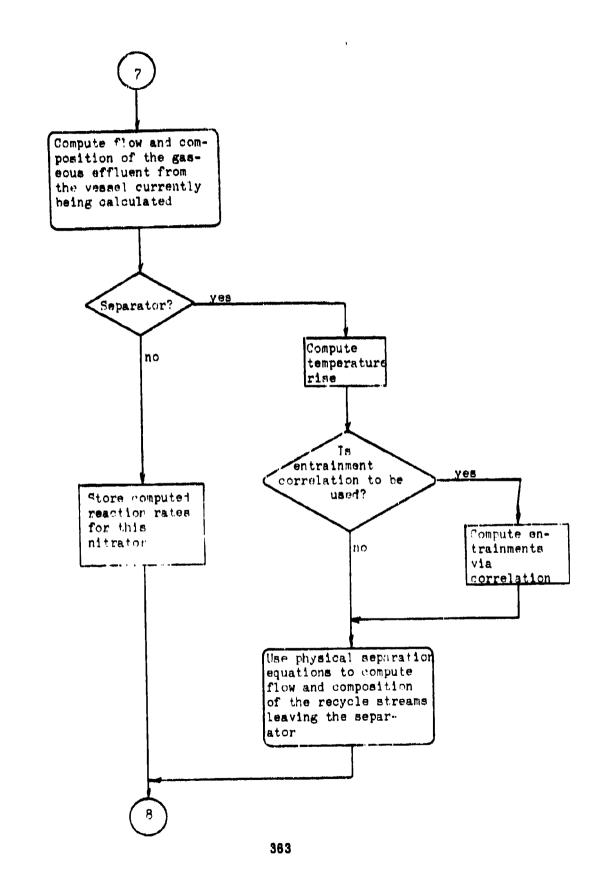


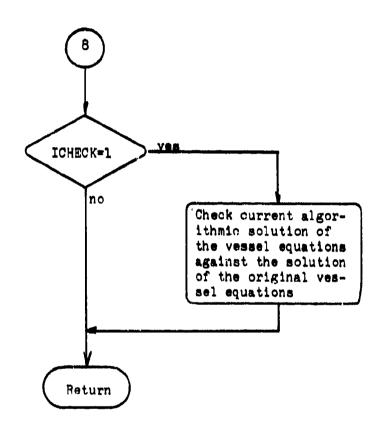


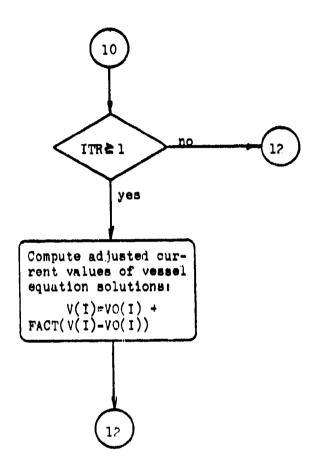


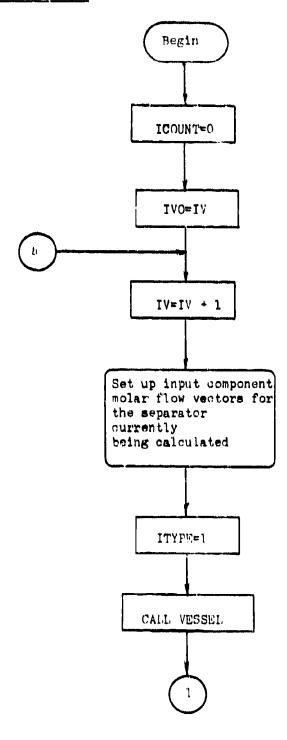
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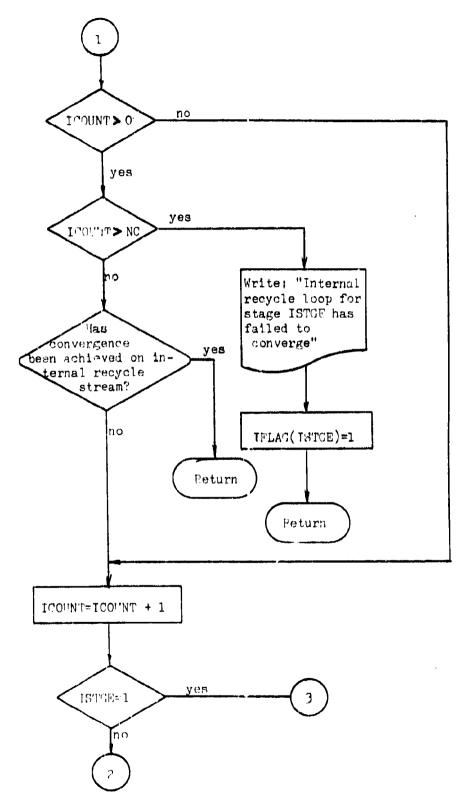


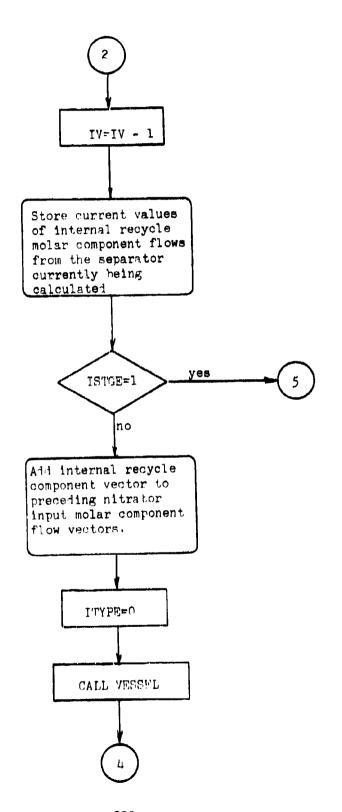


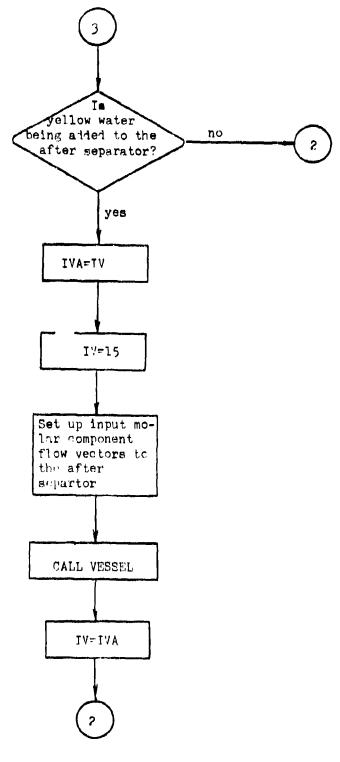


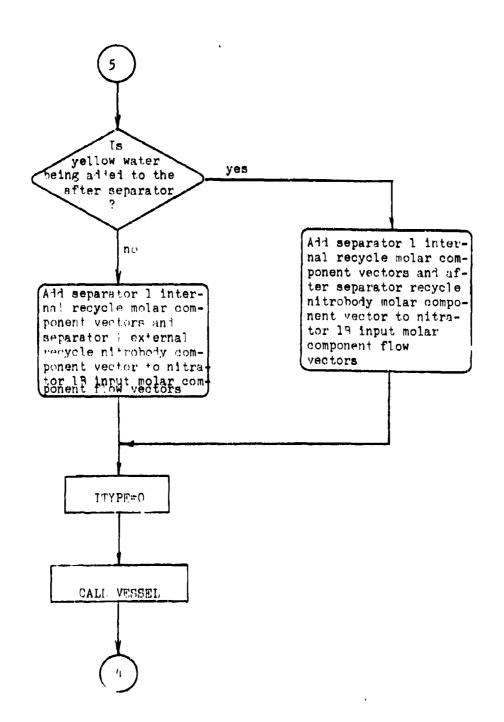


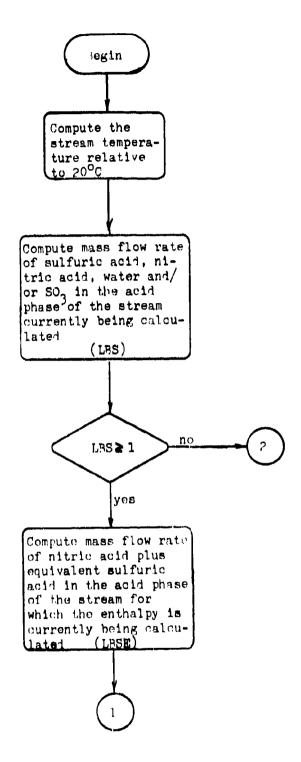


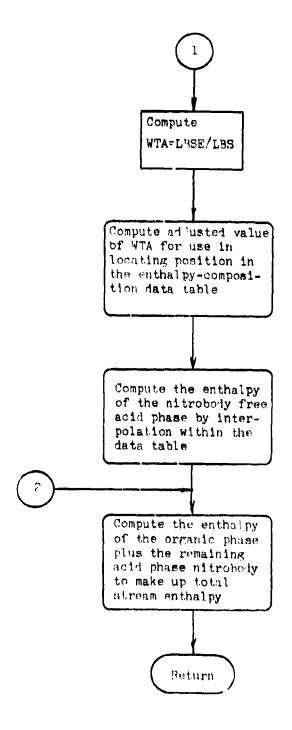












## **APPENDIX** H

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Steady State Simulation--Program Nomenclature

## Main Program

AAS (I)	moles/hr of component i in the acid phase flowing from the after separator
AB	extent of nitration (either $\alpha$ or m) across the stage currently being calculated (moles/hr $NO_2^+$ added to the ring)
AF(I,J)	moles/hr of component i in the entrained acid phase of the organic stream flowing from separator j
AFLBS (I,J)	same as AF but in units of lbs/hr
AF1 (I)	moles/hr of component i in the entrained acid phase of the organic stream flowing from separator 1
AF2 (1)	same as AF1 but for separator 2
AF3 (I)	same as AF1 but for separator 3
AF4 (I)	same as AF1 but for separator 4
AF5 (I)	same as AF1 but for separator 5
AF6(I)	same as AF1 but for separator 6
AI (I)	total acid phase moles/hr of component i flowing into a nitrator which is followed by another nitrator
AII (I)	total acid phase moles/hr (less acid phase of internal recycle) of component i flowing into a nitrator which is followed by a separator
AIR (I,J)	moles/hr of component i in the acid phase of the internal recycle stream flowing from separator j

O	AIR LBS(I,J)	same as AIR but in units of lbs/hr
	AIR1(I)	moles/hr of component i in the acid phase of the internal recycle stream flowing from separator 1
	AIR2(I)	same as AIR1 but for separator 2
	AIR3 (I)	same as AIR1 but for separator 3
	AIR4(I)	same as AIR1 but for separator 4
	AIR5(I)	same as AIR1 but for separator 5
	AIR6 (I)	same as AIR1 but for separator 6
	AKA	general rate factor for all nitration reactions. Also, if Bennett's strong acid rate expression is used, the coefficient of the bisulfate ion concentration
	АКВ	coefficient of the sulfuric acid concentration in Bennett's rate expression
	AKC	coefficient of the pyrosulfuric ion concentration in Bennett's rate expression
	AKEQ	equilibrium constant for the sulfuric acid-water dissociation reaction
	AKEQOV	equilibrium constant for the dissociation of a mixture of sulfuric and pyrosulfuric acids to ionic species. NOT USED

**AKMA** 

AK10

AK12

AKMATL

acid phase mass transfer coefficient

mass transfer coefficient for toluene

ratio of rate constants for reactions 1 & 2

rate constant for reaction 10

AK3S	rate constant for reaction 3 in strong acid
AK3W	rate constant for reaction 3 in weak acid NOT USED
AK4S	rate constant for reaction 4 in strong acid
AK4W	rate constant for reaction 4 in weak acid NOT USED
AK5S	rate constant for reaction 5
AK6S	rate constant for reaction 6
AK7	rate constant for reaction 7
AK8	rate constant for reaction 8
AK9	NOT USED
AN (I)	percent actual nitric acid in the acid phase of the external recycle stream flowing from the ith separator
ANS (I)	mole fraction nitrosylsulfuric acid in the acid phase of the external recycle stream flowing from the i <sup>th</sup> separator
ANS1A	mole fraction nitrosylsulfuric acid in the acid phase flowing from nitrator 1A
ANS3A	mole fraction nitrosylsulfuric acid in the acid phase flowing from nitrator 3A
ARRAY (I,J)	moles/hr of component i in the raw material feedstocks flowing to nitrator j
AS (I)	percent sulfuric acid and SO <sub>3</sub> (as equivalent sulfuric) in the soid phase of the external

TA	total moles/hr of acid phase flowing from the vessel for which the holdup is being computed
АТØМ (I , J )	number of i-type atoms in component j
AV (I, J)	moles/hr of component i in the total acid phase flowing from vessel j
AVI(I,J)	first guess at converged value of moles/hr of component i in the acid phase flowing from vessel j
AXR(I,J)	moles/hr of component i in the acid phase of the external recycle stream flowing from separator j
AXRLBS(I,J)	same as AXR but in units of lbs/hr
AXR1(I)	moles/hr of component i in the acid phase of the external recycle stream flowing from separator 1
AXR2(I)	same as AXR1 but for separator 2
AXR2Ø(I)	previous value of AXR2(I) to which current value is compared during the external recycle convergence procedure
AXR3(I)	same as AXR1 but for separator 3
AXR3Ø(I)	same as AXR2Q bit fpr AXR3
AXR4(I)	same as AXR1 but for separator 4
AXR4Ø (I)	same as AXR2Ø but for AXR4
AXR5(I)	same as AXR1 but for separator 5
AXR5Ø(I)	same as AXR2Ø but for AXR5
AXR6 (I)	same as AXR1 but for separator 6

AXR6Ø (I)	same as AXRIQ but for AXR6
A1A (1)	moles/hr of component i in the total acid phase flowing from nitrator 1A
A1AI (I)	moles/hr of component i flowing into nitrator 1A from the raw material feedstocks
A1B (I)	same as A1A but for nitrator 1B
A1BI (I)	same as A1AI but for nitrator 1B
A18 (I)	moles/hr of component i in the total acid phase flowing from separator 1
A2 (I)	same as A1A but for nitrator 2
A2I (I)	same as A1AI but for nitrator 2
A2S (I)	same as A1S but for separator 2
A3A (I)	same as A1A but for nitrator 3A
A3AI (I)	same as A1AI but for nitrator 3A
A3B (I)	same as A1A but for nitrator 3B
A3BI (I)	same as A1AI but for nitrator 3B
A3S (I)	same as A1S but for separator 3
A4 (1)	same A1A but for nitrator 4
A4I (I)	same as A1AI but for nitrator 4
A4S(I)	same as A1S but for separator 4
A5 (I)	same as A1A but for nitrator 5
A5I (I)	same as A1AI bur for nitrator 5
A5S (I)	same as A1S but for separator 5

A6	<b>(I)</b>
	<b>\-</b> /

same as A1A but for nitrator 6

A61(I)

same as A1AI but for nitrator 6

A6S(I)

same as A1S but for separator 6

BTU(I)

total heat load in nitrator i, BTU/hr

BTUQ(I)

heat evolved in nitrator i which is due to

chemical reaction only, BTU/hr

CDNT

weight & DNT in the nitrobody stream flowing

from separator 6 on and acid free basis

COMP(I,J)

weight % component i in raw material feedstock

type j

CP(I)

heat capacity of component i, BTU/lb-mole/oK

CRUDE

total mass flow rate (lbs/hr) of the organic

stream from separator 6

CSL

cost of oleum, \$/1b

CSN

cost of strong nitric acid, \$/lb

CTF

cost of raw materials per unit of TNT produced,

\$/lb a-TNT

CTL

cost of toluene, \$/lb

CWN

cost of weak nitric acid, \$/lb

DEL

convergence tolerance for components in acid

recycle streams

DH(I)

heat of reaction for reaction i. BTU/lb-mole

of reactant

DIF

storage variable for the difference between

atoms of a given type in and atoms of that type out during material balance calculations

DLT	temperator of separator 1 referenced to 20°C
EAØ (I)	molar ratio of acid phase in the organic stream to total organic stream flowing from the i <sup>th</sup> separator
EAQP(I)	molar ratio of acid phase to organic phase in the organic stream flowing from the ith separator
EFR	enthalpy of mixing for fume recovery acid
EØA (I)	molar ratio of organic phase in the external recycle stream to the total external recycle stream flowing from the ith separator
EØAP(I)	molar ratio of organic phase to acid phase in the external recycle stream flowing from the i <sup>th</sup> separator
EØL	enthalpy of mixing for oleum
EWN	enthalpy of mixing for weak nitric acid
EYW'	enthalpy of mixing for yellow water
E10	activation energy for reaction 10
E21	ratio of activation energy for reaction 2 to that of reaction 1
F3S	activation energy for reaction 3
E3W	NOT USED
E4S	activation energy for reaction 4
E4W	NOT USED
E5S	activation energy for reaction 5
E8S	activation energy for reaction 6

1.7	activation energy for reaction 7
E8	activation energy for reaction 8
E9	NOT USED
FA	volume fraction of acid phase in the vessel for which holdup is being calculated
FACD(I)	NOT USED
FACT2	relaxation factor used in convergence of the external recycle from separator 2
FACT3	same as FACT2 but for separator 3
FACT4	same as FACT2 but for separator 4
FACT5	same as FACT2 but for separator 5
FACT6	same as FACT2 but for separator 6
FD	extent of reaction for the decomposition of nitrosyleulfuric acid
FDN	moles/hr of ring associated NO <sub>2</sub> groups flowing into a nitration stage with the organic stream from the previous stage
FIN (I,J)	lbs/hr of feedstock i to nitration vessel j
FMAX	maximum possible volumetric flow rate of material through any internal recycle pipe
FØ	volume fraction of organic phase in the vessel for which holdup is being calculated
FØV	NOT USED

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**GA (I,J)** 

number of i-type atoms in gaseous product j

CGS (I,J)	moles/hr of gaseous product j evolved in vessel i
G1 (I)	ratio of the Arrhenius coefficients in the kinetic rate expressions for reactions 1 and 2 in the i <sup>th</sup> nitrator
G10 (I)	Arrhenius coefficient for reaction 10 in nitrator i
G15 (I)	moles/hr of CO <sub>1.8</sub> evolved from vessel i
G16 (I)	moles/hr of $NO_{\chi}$ evolved from vessel i
G18 (I)	moles/hr of TNM evolved from vessel i
G19 (I)	moles/hr of CO <sub>2</sub> evolved from vessel i
G3 (I)	NOT USED
G3S (I)	Arrhenius coefficient for reaction 3 in nitrator i
G4 (I)	NOT USED
G4S (I)	same as G3S but for reaction 4
G5 (I)	same as G3S bur for reaction 5
G6 (I)	same as G3S but for reaction 6
G7 (I)	same as G3S but for reaction 7
G8 (I)	same as G3S but for reaction 8
G9 (I)	Zero
HDR (I,J)	array used to store alphanumeric headings for chemical elements in material balance printout
I	general index integer
IAFT	after separator switch. If equal to 1 then after separator is considered in the simulation as a dilution vessel to which 1000 lb/hr of yellow water is added

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Ü	ICHECK	check calculation switch. If equal to 1 then check calculations are performed on the vessel equations in SUBROUTINE VESSEL
	IE	entrainment correlation switch. If equal to 0 then entrainment correlation is used instead of assuming fixed values for entrainments
	IFLG (I)	set equal to 1 whenever internal recycle loop for stage i does not converge
	11	index used in calculation of extent of nitration in each stage
	III	number of passes through SUBROUTINE VESSEL required to reach a converged solution
	IJ	index used in calculation of extent of nitration in each stage
	ILIST	printout switch. If equal to 1 printout of phase compositions, gas flows, vessel time constants, volumetric flows, acid phase solubility, vessel holdups and stream compositions is obtained
	IMBAL	material balance switch. If equal to 1 chemical element balances are carried out on the entire nitration section, each individual nitration stage, and each individual vessel
	IN	nitrator number
	INIT (I)	vessel index. Has value of 1 if vessel is a nitrator

IPI

and 0 if vessel is a separator

intermediate printout switch. If equal to 1 a printout of external recycle molar component flows after each iteration will be given

IPNCH	punch switch. If equal to zero no punched output. If equal to 1 final steady state (i.e. values of component molar flows for both phases in each vessel) is punched out for use as initial conditions in dynamic simulation or new first guess in steady state simulation
IPRNT	SUBROUTINE VESSEL printout switch. If equal to 1 gives a variety of debug printout which can be used to analyze problems associated with the vessel equations
ISTGE	stage number
ISTØP	equal to number of data sets plus 1. Program execution is terminated when NSET = $ISTQP$
ITYPE	equal to 0 for a nitrator and 1 for a separator
IV	vessel number
IV1	vessel number used during initial determination of recycle streams from first guess values of component molar phase flows
IX	index used in calculation of extent of nitration in each stage
IY	index used in calculation of extent of nitration in each stage
J	general index integer used for a variety of counting purposes
К	general index integer used for a variety of counting purposes
KEY	index which indicates whether extent of alpha or meta nitration is to be determined in a given stage
L	vessel index used in holdup calculation printout

M	vessel index used in holdup calculation printout
MHQLDA	total moles of acid phase holdup in the vessel currently being calculated
MHQLDQ	total moles of organic phase holdup in the vessel currently being calculated
MW (I)	molecular weight of component i
NATM	index of atomic species used in elemental material balance relculations
NC	maximum number of iterations allowed on any given recycle (internal or external) convergence loop
NC1	running sum of iterations on external recycle loop for separator 2
NC 2	running sum of iterations on external recycle loop for separator 3
NC3	running sum of iterations on external recycle loop for separator 4
NC4	running sum of iterations on external recycle loop for separator 5
NC5	running sum of iterations on external recycle loop for separator 6
NDS	number of sets of operating conditions for which r steady state solution is desired in a given simulation run
N <b>Ø</b> (I)	notch setting on the internal recycle gate valve for separator i
NSET	number of the data set for which the steady state solution is currently being determined

NTOS (I)	molar ratio of nitric acid to the sum of sulfuric acid and SO; in the external recycle stream flowing from the ith separator
NTØS1A	molar ratio of nitric acid to the sum of sulfuric acid and SO <sub>3</sub> in the acid phase flowing from nitrator 1A
NTØS3A	same as NTOS1A but for nitrator 3A
<b>QUTN</b>	moles/hr of ring associated NO; groups flowing out of a given nitration stage
PACID(I,J)	pounds of component i held up in the acid phase of vessel j
PAS (I)	moles/hr of component i in the organic phase flowing from the after separator
PCTAS	weight % dissolved nitrobody in the acid phase flowing from the after separator under the condition of 1000 lb/hr of yellow water added to the after separator
PCTD	weight % DNT in the organic components dissolved in the acid phase flowing from separator 1
PCTNB	weight % total nitrobody contained in the acid stream flowing from separator 1 to the after separator
PCTNBD	weight % dissolved nitrobody contained in the acid stream flowing from separator 1 to the after separator
PF (I,J)	moles/hr of component i in the organic phase of the organic stream flowing from separator j

PFLBS(I,J)

same as PF but in units of lb/hr

)	PF1(I)	moles/hr of component i in the organic phase of the organic stream flowing from separator 1
	PF2(I)	same as PFJ but for separator 2
	PF3(I)	same as PF1 but for separator 3
	PF4(I)	same as PF1 but for separator 4
	PF5 (I)	same as PF1 but for separator 5
	PF6 (I)	same as PF1 but for separator 6
	PI (I)	total organic phase moles/hr of component i flowing into a nitrator which is followed by another nitrator
	PII (I)	total organic phase moles/hr of component i (less organic phase of internal recycle) flowing into a nitrator which is followed by another separator
	PIR (I,J)	moles/hr of component i in the organic phase of the internal recycle stream flowing from separator j
	PIRLBS (I,J)	same as FIR but in units of 1b/hr
	PIR1 (I)	moles/hr of component i in the organic phase of the internal recycle stream flowing from separator 1
	PIR2(I)	same as PIR1 but for separator 2
	PIR3(I)	same as PIR1 but for separator 3
	PIR4 (I)	same as PIR1 but for separator 4
	PIR5 (I)	same as PIR1 but for separator 5
	PIR6 (I)	same as PIR1 but for separator 6

PNS	weight % nitrosylsulfuric acid in the acid phase of the external recycle stream flowing from a given separator
PØUNDS (I,J)	pounds of component i held up in the organic phase of vessel j
PT	total moles/hr of organic phase flowing from the vessel for which the holdup is being computed
PV (I,J)	moles/hr of component i in the total organic phase flowing from vessel j
PVI (I,J)	first guess at converged value of moles/hr of component i in the organic phase flowing from vessel j
PXR(I,J)	moles/hr of component i in the organic phase of the external recycle stream flowing from separator j
PXRLBS (I,J)	same as PXR but in units of lb/hr
PXR1(I)	moles/hr of component i in the organic phase of the external recycle street flowing from separator 1
PXR2(I)	same as PXR1 but for separator 2
PXR2Ø(I)	previous value of PXR2(I) to which current value is compared during the external recycle convergence procedure
PXR3(I)	same as PXR1 but for separator 3
PXR3Ø(I)	same as PXR2Q but for separator 3
PXR4 (I)	same as PXR1 but for separator 4
PXR4Ø(I)	same as PXR2Ø but for separator 4
PXR5(I)	same as PXR1 but for separator 5
PXR5Ø(I)	same as PXR2Ø but for separator 5

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PXR6 (I)	same as PXR1 but for separator 6
PXR00(I)	same as PXR20 but for separator 6
P1A (I)	moles/hr of component i in the total organic phase flowing from nitrator 1A
P1AI	moles/hr toluene to nitrator 1A
P1B (I)	same as P1A but for nitrator 1B
P1BI	moles/hr toluene to nitrator 1B
P1S (I)	moles/hr of component i in the total organic phase flowing from separator 2
P2 (I)	moles/hr of component i in the total organic phase flowing from nitrator 2
P2S (I)	same as P1S but for separator 2
P3A (I)	same as P2 but for nitrator 3A
P3B (I)	same as P2 but for nitrator 3B
P3S (I)	same as P1S but for separator 3
P4 (I)	same as P2 but for nitrator 4
P48 (I)	same as P1S but for separator 4
P5 (I)	same as P2 but for nitrator 5
P5S (I)	same as P1S but for separator 5
P6 (I)	same as P2 but for nitrator 6
P6S(I)	same as P1S but for nitrator 6
Q	accumulation term used in calculation of nitrator heat loads

QA	ft <sup>3</sup> /hr of total acid phase flowing from the vessel for which holdup is being computed
QP	same as QA but for organic phase
QQ(I)	fraction nitric acid converted to nitronium ion in vessel i
QT	ft <sup>2</sup> /hr of total material flowing from the vessel for which holdup is being computed
R (I)	rate of reaction for reaction i in the vessel currently being simulated by SUBROUTINE VESSEL
RATES (I, J)	rate of reaction for reaction j in nitrator i
RECN	moles/hr of ring associated NO <sub>2</sub> groups flowing into a nitration stage with the external recycle from the next higher stage
RH <b>Q</b> (I)	molar density of component i
RHØA	molar density of the acid phase in the vessel for which holdup is being calculated
rhønbs (I)	mass density of the nitrobody dissolved in the acid phase of the external recycle stream flowing from the i <sup>th</sup> separator
RHØP	molar density of the organic phase in the vessel for which holdup is being calculated
RHØ1	equivalent to RHØ (1)
RHØ10	equivalent to RHØ (10)
RHØ11	equivalent to RHQ (11)
RHQ12	equivalent to RHQ(12)
RHØ13	equivalent to RHQ (13)

RHØ14	equivalent to RHØ (14)
RHQ2	equivalent to RHØ(2)
<b>RHØ3</b>	equivalent to RHØ(3)
RHQ4	equivalent to RHØ(4)
RHQ5	equivalent to RHØ(5)
RHQ6	equivalent to RHQ (6)
RHØ7	equivalent to RHG (7)
RHØ8	equivalent to RHQ(8)
RHQ9	equivalent to RHØ(9)
RP	coefficient of the molecular oxygen term in reaction 9
RPP	precomputed constant based on RP which is used in material balance calculations
RR	ideal gas law constant
SA	adjustable constant. NOT USED
SB	equivalent to KMAP
SC	equivalent to AKEQ4 in SUBROUTINE VESSEL
SD	adjustable constant. NOT USED
SE	equivalent to AK9. NOT USED
SG	equivalent to E9. NOT USED
SH	equivalent to AKEQ1 in SUBROUTINE VESSEL
SI	

SJ	equivalent to AKEQ3 in SUBROUTINE VESSEL
SNA(I)	net moles/hr of ring associated alpha NO <sub>2</sub> groups flowing through the i <sup>th</sup> nitration stage. Extent of alpha-nitration in nitration stage i
SNM (I)	same as SNA but for meta-nitration
SOL(I)	saturation solubility of total organic components in the acid phase of vessel i
SPA	mass flow rate of the acid phase of the external recycle stream (spent acid) from separator 1
SPACID	mass flow rate of the total external recycle stream from separator 1
SPP	mass flow rate of the organic phase of the external recycle stream from separator 1
STN	total flow rate (lb/hr) of strong nitric acid into the process from all raw material feed streams
SULP	total flow rate (lb/hr) of cleum into the process from all raw material feed streams
SUM	total molar flow rate of gasses evolved in the nitration stage for which material balance calculations are being executed. Also the moles/hr of organics flowing from separator 6 with the organic stream
SUMA	mass flow rate acid components of the acid phase from the after separator when the after separator option is used
SUMI	accumulation term for input streams used during material balance calculations (moles/hr)
SUMØ	mass flow rate of organic components in the acid phase from the after separator when the after separator option is used

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S15	total moles/hr of $CO_{1,\delta}$ evolved in nitration section
S16	total moles/hr of $NO_{\chi}$ evolved in the nitration section
S18	total moles/hr of TNM evolved in the nitration section
S19	total moles/hr of CO <sub>2</sub> evolved in the nitration section
TANB	total moles/hr of dissolved organics in the acid phase of the external recycle flowing from separator 1
TAS(I)	total acidity of the acid phase of the external recycle stream flowing from separator i
TATMI	total moles/hr of a given atomic specie entering that section of the process over which an elemental material balance is being computed
ТАТМ <b>Ø</b>	total moles/hr of a given atomic specie exiting that section of the process over which an elemental material balance is being computed
TAU	holdup time in a given vessel
TCØX	equivalent to \$15
TCO2	equivalent to \$19
TDNT	total mass flow rate of DNT in the organic stream from separator 6
TDNTA	total pounds of DNT held up in the acid phase of all process vessels
TDNTP	total pounds of DNT held up in the organic phase of all process vessels

TEMP(I) temperature of the ith nitrator

TITLE (I) alphanumeric identifying information for a given

data set

TLBS total lb/hr of acid components in the acid phase

of the external recycle stream flowing from the separator for which acid composition calcula-

tions are being executed

TMQLES same as TLBS but in units of moles/hr

TN(I) weight % total nitric acid in the acid phase of the

external recycle stream flowing from the ith

separator on an organic free basis

TNØX equivalent to \$16

TNTI moles/hr of TNT entering the nitration section

with feed material

TOL lb/hr toluene fed to the nitration section

TPNB total moles/hr of organic components in the

organic phase of the external recycle stream

flowing from separator 1

TPØUND accumulation term for total pounds of organic

material held up in either the acid or organic phase of the vessel for which holdup calculations

are being executed

TR reference temperature

TS(I) weight % total sulfuric acid plus SO<sub>3</sub> (as equivalent

sulfuric) in the acid phase of the external recycle stream flowing from the ith separator on an organic

free basis

TSEP(I) temperature of the ith separator

TSEP1 equivalent to TSEP (1)

TSEP2 equivalent to TSEP (2)

TSEP3 equivalent to TSEF (3)

TSEP4 equivalent to TSEP (4)

TSEP5 equivalent to TSEP (5)

TSEP6 equivalent to TSEP (6)

TTDNT total pounds of DNT held up in all process vessels

TTLBS (I) total pounds of organic phase held up in the

ith vessel

TTLBSA(I) total pounds of organic components held up in

the acid phase of the ith vessel

TTNM equivalent to \$18

TTNTA total pounds of TNT held up in the acid phase

of all process vessels

TTNTP total pounds of TNT held up in the organic phase

of all process vassels

TTTLBA total pounds of organic components held up in

the acid phase of all process vessels between

and including nitrator 2 and separator 6

TTTLBS total pounds of organic phase held up in all

process vessels between and including nitrator 2

and separator 6

TTTNT total pounds of TNT held up in all process vessels

TTTTLB total pounds of organic components (in both acid

and organic phases) held up in all process vessels

between and including nitrator 2 and separator 6

TVØL volumetric flow of the organic compounds in the

acid phase of the external recycle stream flowing from a separator for which nitrobody density is

being computed

T1A	temperature of nitrator 1A
T1B	temperature of nitrator 1B
T2	temperature of nitrator 2
ТЗА	temperature of nitrator 3A
тзв	temperature of nitrator 3B
T4	temperature of nitrator 4
T5	temperature of nitrator 5
T6	temperature of nitrator 6
V (I)	storage vector for current values of the i component flow terms generated in SUBROUTINE VESSEL
VHQLDA	volume of acid phase held up in a given vessel
VHØLDØ	volume of organic phase held up in a given vessel
VKM (I)	working volume of the ith nitrator
VKMS	working volume of any separator
VØL(I)	working volume of the ith vessel
WKN	total flow rate (lb/hr) of weak nitric acid into
	the process from all raw material feed stream
XP (I, J)	the process from all raw material feed stream mole fraction of component i in the organic phase within vessel j
XP (I, J)  XPA (I, J)	mole fraction of component i in the organic

**YIELD** 

moles  $\alpha$  -TNT in the organic stream flowing from separator 6 per mole of toluene fed to the process

(expressed as %)

ZA

temperature coefficient in the exponent of the Arrhenius rate law

### Subroutine NITSEP

The definitions of the following variables are identical to those in the main program: AF(I,J), AF1(I), AI(I), AII(I), AIR(I,J), AIR1(I), AV(I,J), AXR(I), AXR1(I), DEL, EAØ(I), EAØP(I), EØA(I), EØAP(I), FACD(I), G15(I), G18(I), G18(I), G19(I), I, IAFT, ICHECK, IE, IFLG(I), IN, IPRNT, ISTGE, ITYPE, IV, NØ(I), PF(I,J), PF1(I), PI(I), PII(I), PIR(I,J), PIR1(I), PV(I,J), PXR1(I), RATES(I,J), SØL(I), TSEP(I), YELLOW(I),

### Remaining variables are defined as follows:

AIRØ(I) previous value of AIR(I,J) to which current

value is compared during the internal recycle

convergence procedure for stage j

ICOUNT counter for iterations of internal recycle

convergence loop

IVA vessel index for separator 1 used whenever

after separator is included in the simulation

IVØ vessel index for the nitrator which preceeds

the separator for which internal recycle

convergence is being executed

PIRQ(I) previous value of PIR(I,J) to which current

value is compared during the internal recycle

convergence procedure for stage j

### Subroutine VESSEL

The definitions of the following variables are identical to those in the main program: AF (I,J), AI (I), AIR (I,J), AKA, AKEQ, AKEQOV, AKMA, AKMAP, AKMATL, AK10, AK12, AK3S, AK3W, AK4S, AK4W, AK4S, AK6S, AK7, AK8, AV (I,J), AXR (I,J), CP (I), DEL, DH (I), EAØ (I), EAØP (I), EØAP (I), E10, E12, E3S, E3W, E4S, E4W, E5S, E6S, E7, E8, FACD (I,J), FP, FMAX, G15 (I), G16 (I), G18 (I), G19 (I), I, IAFT, ICHECK, IE, III, IN, IPRAT, ISTGE, ITYPE, IV, J, MN (I), NØ (I), PF (I,J), PI (I), PIR (I,J), PV (I,J), PXR (I,J), QQ (I), R (I), RATES (I,J), RHØ1, RHØ10, RHØ12, RHØ13, RHØ14, RHØ2, RHØ3, RHØ4, RHØ5, RHØ6, RHØ7, RHØ6, RHØ9, RP, RR, SA, SB, SC, SD, SE, SG, SH, SI, SJ, SØL (I), TEMP (I), TR, TSEP (I), V (I), VKM (I), VKMS, YELLØW (I)

The definitions of the following variables are identical to other variables defined in the main program:

Variable in VESSEL	Variable in main program
All thru All7	<b>A</b> I(1) thru AI(17)
F1(I) thru F10(I)	G1(I) thru G10(I)
ISEP	ISTGE
PI1 thru PI17	PI(1) thru PI(17)
R1 thru R10	R(1) thru R(10)

# Remaining variables are defined as follows:

A (I)	moles/hr of component i in the acid phase flowing from the vessel currently being simulated
AINB	moles/hr of organic components in the combined acid phase flowing to the vessel currently being simulated
AKEQ1	equilibrium constant for the dissociation of nitric acid in aqueous sulfuric acid
AKEQ2	equilibrium constant for the dissociation of sullfuric acid in an aqueous system
AKEQ3	equilibirum constant for dissociation of nitric acid in anhydrous sulfuric acid

AKEQ4	equilibrium constant for the recombination of nitronium and bisulfate ions in anydrous acid mixtures
AK1	computed value of AKEQ1 obtained from interval halving procedure
AK3	computed value of AKEQS obtained from interval halving procedure
AK4	computed value of AKEQ4 obtained from interval halving procedure
AL	moles/hr water in the acid phase flowing from the vessel currently being simulated; negative values represent moles/hr SO <sub>3</sub>
AM (I)	moles/hr of component i transferred from the organic phase into the acid phase by bulk mass transfer in the vessel currently being simulated. Used in check calculations only
AMNB	total moles/hr of organic components transferred from the organic to acid phase by bulk mass transfer in the vessel currently being simulated
AM1	moles/hr $\alpha$ -MNT transferred from the organic phase into the acid phase by bulk mass transfer in the vessel currently being simulated
ANB	moles/hr organic components in the acid phase flowing from the vessel currently being simulated
ASØ4	combined moles/hr of $\rm H_2SO_4$ and $\rm SO_3$ in the acid phase flowing from the vessel currently being simulated
AT	total moles/hr of acid phase flowing from the vessel currently being simulated
ATI	reciprocal of AT

AXL	same as AL but used only in check calculations
AX1	1. + C1A-CA
AX1 thru AX9	same as A1 thru A9 but used only in check calculations
AX12	same as A12 but used only in check calculations
A1	moles/hr $\alpha$ MNT in the acid phase flowing from the vessel currently being simulated
A2	same as A1 but for m MNT
A3	same as A1 but for $\alpha$ DNT
A4	same as A1 but for m MNT
A5	same as A1 but for $\alpha$ TNT
A6	same as A1 but for m TNT
A7	same as A1 but for TNBX
A8	same as A1 but for TNB
A9	moles/hr nitric acid in the acid phase flowing from the vessel currently being simulated
A11	same as A9 but for H <sub>2</sub> SO <sub>4</sub>
A12	same as A9 but for nitrosylsulfuric acid
A13	same as A9 but for water
A14	same as A9 but for SO <sub>3</sub>
A17	moles/hr nitronium ion in the acid phase flowing from the vessel currently being simulated
В	free sulfuric acid concentration in oleum in the vessel currently being simulated

вс	B + (3 x C)
B1 .	term representing moles/hr of an organic component flowing into a vessel with the combined acid phase plus the amount of that component formed by reaction
B2	moles/hr of an organic component flowing into a vessel with the combined organic phase
С	concentration of pyrosulfuric acid in the vessel currently being simulated
CA	ratio of AMNB to ANB (constant used in determining bulk mass transfer during simulataneous solution of vessel equations)
СВ	$(.5 \times B) + (1.5 \times C)$
CF	C - F
CHNØ3	equilibrium concentration of nitric acid in the acid phase of the vessel currently being simulated (moles/cf <sup>2</sup> )
CHSØ4	same as CHNØ3 but for bisulfate ion
CH <b>S2</b> Ø7	same as CHNØ3 but for pyrosulfate ion
CH2Ø	same as CHNØ3 but for water
CH28O4	same as CHN93 but for sulfuric acid
СНЗФ	same as CHNØ3 but for hydronium ion
CNØ2	same as CHNØ3 but for nitronium ion
CR	ratio of AMNB to PNB. Constant used in determination of bulk mass transfer during simultaneous solution of vessel equations

computed volumetric flow of an internal recycle stream based on entrainment correlation

CVØL

CXX	correlation used to determine fraction organic entrained in the internal recycle stream
CX2	ratio of fraction internal recycle to fraction external recycle used in entrainment correlation
01	equal to overall mass transfer coefficient (ETA) during simultaneous solution of vessel equations
C1A	coefficient equal to C1/ANB used during simultaneous solution of vessel equations
C1Ø	coefficient equal to C1/PNB used during simul- taneous solution of vessel equations
C1P	coefficient related to toluene diffusion which is used during simultaneous solution of vessel equations
C11	coefficient equal to acid phase mass transfer plus the sum of the reaction coefficients used during simultaneous solution of the vessel equations
C12	coefficient equal to (~ CIØ - CR) used during simultaneous solution of the vessel equations
C21	coefficient equal to (CA - C1A) used during simultaneous solution of the vessel equations
C22	coefficient equal to $(1 + C10 + CR)$ used during simultaneous solution of the vessel equations
C4	equal to the sum of reaction rates R1 thru R6. Represents the moles/hr water produced by reactions 1 thru 6
C5	moles/hr nitrosylsulfuric acid produced by oxidation reactions
C5P	net moles/hr nitrosylsulfuric acid produced after decomposition takes place

C6	organic phase nitric acid solubility coefficient
DD (I)	rate of diffusion of organic component i. Used only in check calculations
DLA	convergence tolerance for components in both acid and organic phases flowing from the vessel currently being simulated
DRM	difference in molar densities between the acid and organic phases used in the entrainment correlation
D1	moles/hr of α MNT which diffuse from the organic into the acid phase in the vessel currently being simulated. Negative values means diffusion is from acid to organic phase
D10	moles/hr of toluene which diffuse from the bulk organic phase to the acid-organic interface
D2	same as D1 but for mMNT
D\$	same as D1 but for a DNT
D4	same as D1 but for mDNT
D5	same as D1 but for a TNT
D6	same as D1 but for mTNT
מ7	same as D1 but for TNBX
D8	same as D1 but for TNB
BAG4	molar ratio of acid phase in the organic stream to total organic stream, based on the entrainment correlation
ETA	overall mass transfer coefficient in the vessel currently being simulated

ETAA	acid phase mass transfer coefficient in the version currently being simulated
ETAP	organic phase mass transfer coefficient in the vessel currently being simulated
BTATL	mass transfer coefficient for toluene in the organic phase
EXX	ratio acid phase to organic phase in the organic stream, based on the entrainment correlation
F	initial concentration of nitric acid prior to establishment of equilibrium conditions in the vessel currently being simulated
FA	volume fraction acid phase in the vessel currently being simulated
FACT	relaxation factor used in convergence procedure for composition of phases flowing from the vessel currently being simulated
FAR	fraction of the total acid phase flowing from a separator that is recycled for the separator currently being simulated
FAR1	fraction of the total acid phase flowing from a separator that is entrained in the organic stream which flows to the next nitrator for the separator currently being simulated
FC	constant equal to (2 x F) - C used in computation of nitronium ion equilibrium concentration in region 2 for the vessel currently being simulated
FF (I)	cross sectional area fraction corresponding to the ith notch setting on the gate valve in the internal recycle line of the separator currently being simulated

FHG upper boundary value of internal recycle fraction computed using the entrainment coefficient FIR internal recycle fraction computed using the entrainment correlation for the separator currently being simulated FLW same as FHG except lower boundary value molar ratio of organic phase in the acid recycle FOG streams to total organic phase flowing from the separator currently being simulated (based on entrainment correlation) FOGC current value of FOG FP volume fraction of organic phase in the vessel currently being simulated FPO molar fraction of the total organic phase flowing from a separator which appears in the organic stream for the separator currently being simulated FPØ1 molar fraction of the total organic phase flowing from a separator which appears in the recycled acid streams for the separator currently being simulated FR volume fraction of the combined recycled acid streams which is external recycle for the separator currently being simulated FRAC volume fraction of the combined recycled acid

)

**FVKM** volume of the vessel currently being simulated

**GAMMA** acid concentration factor for all nitration reactions in the vessel currently being simulated

G1	equivalent to F1 (I) where I is the index of the nitrator currently being simulated or the index of the nitrator preceding the separator currently being simulated
G10	same as G1 but for F10(I)
G10P	rate of reaction 10 per mole of TNT in the acid phase flowing from the vessel currently being simulated
G15P	equivalent to G15(I) where I is the index of the vessel currently being simulated
G3	same as G1 but for F3(I)
G3D	rate of reaction 3 per mole of $\alpha$ MNT in the acid phase flowing from the vessel currently being simulated
G3S	same as G1 but for F3S(I)
G4	same as G1 but for F4(I)
G4D	rate of reaction 4 per mole of mMNT in the acid phase flowing from the vessel currently being simulated
G48	same as G1 but for F4S(I)
<b>G</b> 5	same as G1 but for F5(I)
G5G	rate of reaction 5 per mole of $\alpha$ DNT in the acid phase flowing from the vessel currently being simulated
G6	same as G1 but for F8(I)
G6G	rate of reaction 6 per mole of mDNT in the acid phase flowing from the vessel currently being simulated

G7	same as G1 but for F7(I)
G7G	rate of reaction 7 per mole of DNT in the acid phase flowing from the vessel currently being simulated
G8	same as G1 but for F8(I)
G8P	rate of reaction 8 per mole of DNT in the acid phase flowing from the vessel currently being simulated
G9	same as G1 but for F9(I)
G9G	zero
ICØUNT	number of passes made through sucroutine VESSEL during a particular convergence iteration for the vessel currently being simulated
ITR	number of convergence iterations executed for the vessel currently being simulated. Five iterations, each consisting of NC passes through VESSEL are allowed
MWAVGA	average molecular weight of the acid phase flowing from the separator which is being simu- lated and for which entrainment is being determ- ined via the correlation
MWAVGP	same as MWAVCA but for the organic phase
NC	maximum number of passes through VESSEL allowed per iteration during vessel equation convergence procedure

value of N( ,I) where I is the index of the separator currently being simulated

NØTCH

P (I)

moles/hr of component i in the organic phase flowing from the vessel currently being simulated

Вид	moles/hr of combined organic components in the organic phase flowing from the vessel currently being simulated
PP	exponent of the nitrobody-in-acid solubility correlation
PT	moles/hr of combined organic components in both the acid and organic phases flowing from the vessel currently being simulated
PTI	the reciprocal of PT. Used for computation of entrainment via the correlation
PX (I)	same as P(I) for components 1 thru 8 but used only in check calculations
PX10	same as P(10) but used only in check calculations
PX9	same as P(9) but used only in check calculations
P1	moles/hr $\alpha$ MNT in the organic phase flowing from the vessel currently being simulated
P10	same as P1 but for toluene
P2	same as P1 but for mMNT
P3	same as P1 but for a DNT
P4	same as P1 but for mDNT
P5	same as P1 but for a TNT
P6	same as P1 but for mTNT
P7	same as P1 but for TNBX
P8	same as P1 but for TNB
P9	same as P1 but for nitric acid

Q	fraction of molecular nitric acid converted to nitronium ion in the vessel currently being simulated
QA	ft <sup>8</sup> /hr of acid phase flowing from the vessel currently being simulated
QP	ft <sup>2</sup> /hr of organic phase flowing from the vessel currently being simulated
QT	volumetric flow (ft <sup>3</sup> /hr) of combined internal plus externs' recycle streams flowing from the separator currently being simulated
RHLØ	mass density (lb/ft <sup>\$</sup> ) of the organic phase flowing from the separator currently being simulated and for which entrainment is being determined via the correlation
RHQQ	same as RHLØ but molar density (moles/ft <sup>3</sup> )
RR1	same as R1 but used only for check calculation
RR10	same as R10 but used only for check calculation
RR3	same as RR1 but for R3
RR7	same as RR1 but for R7
RRS	same as RR1 but for R8
RR9	same as RR1 but for R9
RR9G	same as RR1 but for R9G
RTL	mass ratio of organic to acid phase in the separator currently being simulated and for which entrainment is being calculated via the correlation
RTØ	same as RTL but molar ratio

RVQL	maximum volumetric flow of internal recycle stream possible for a given notch setting in the separator currently being simulated and for which entrainment is being calculated via the correlation	
R10A	rate of reaction 10 for α TNT in the vessel currently being simulated	
R10M	rate of reaction 10 for mTNT in the vessel currently being simulated	
R7A	rate of reaction 7 for mDNT in the vessel currently being simulated	
R8A	rate of reaction 8 for a DNT in the vessel currently being simulated	
R8M	rate of reation 8 for mDNT in the vessel currently being simulated	
R9G	zero	
R9GA	zero	
R9GM	zero	
8	initial concentration (moles/ft <sup>3</sup> ) of total sulfuric acid in the acid phase of the vessel currently being simulated	
SANB	same as ANB but used only in check calculations	
SANBC	same as SANB but based on computed solubility	
SPNB	same as PNB but used only in check calculations	
SUM	sum of the individual organic component diffus- sions used only in the check calculations	
TAF	moles/hr of entrained acid phase (based on the correlation) contained in the organic stream flowing from the separator currently being simulated	

Ü	TAIR	moles/hr of acid phase (based on the correlation) contained in the internal recycle stream flowing from the separator currently being simulated
	TAR	molus/hr of acid phase (based on the correlation) contained in the combined internal and external recycle streams flowing from the separator currently being simulated
	TAXR	moles/hr of acid phase (based on the correlation) contained in the external recycle stream flowing from the separator currently being simulated
	TKEL	temperature (in Degrees Kelvin) of the vessel currently being simulated
	TLI	total mass flow (lb/hr) through the separator currently being simulated and for which entrainment is being computed via the correlation
	TPF	moles/hr of organic phase (based on the correlation) contained in the organic stream flowing from the separator currently being simulated
	TPIR	moles/hr of entrained organic phase (based on the correlation) contained in the internal recycle stream flowing from the separator currently being simulated
	TPR	moles/hr of entrained organic phase (based on the correlation) contained in the combined recycle streams flowing from the separator currently being simulated
	TPXR	moles/hr of entrained organic phase (based on the correlation) contained in the external recycle stream flowing from the separator currently being simulated

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<b>VØ</b> (I)	storage vector for the values of the i component molar flows generator on the previous pass through VESSEL for the vessel currently being simulated
W	concentration (moles/ft <sup>2</sup> ) of water in the vessel currently being simulated
x	computed equilibrium concentration of nitronium ion in the acid phase under aqueous conditions and under conditions of cleum region 3. Also equilibrium concentration at molecular nitric acid in cleum region 2
XA	NOT USED
хв	NOT USED
xc	NOT USED
AD	NOT USED
XE	NOT USED
XEQA	equilibrium solubility of organic material (nitrobody) in the acid phase of the vessel currently being simulated
XF	NOT USED
XL	lower boundary value used in an interval halving convergence procedure
<b>χ</b> υ	upper boundary valued used in an interval halving convergence procedure
X1	mole fraction α MNT in the acid phase of the vessel currently being simulated
X11	same as X1 but for sulfuric acid
X12	same as X1 but for nitrosylsulfuric acid

X13	same as X1 but for water
X14	same as X1 but for SO <sub>8</sub>
X2	same as X1 but for mMNT
жs	same as X1 but for a DNT
X4	same as X1 but for mDNT
<b>X</b> 5	same as X1 but for $\alpha$ TNT
<b>X6</b>	same as X1 but for mTNT
ж7	same as X1 but for TNBX
X8	same as X1 but for TNB
X9	same as X1 but for nitric acid
Y	equilibrium concentration of the pyrosulfate ion in the acid phase of the vessel being simulated under conditons of oleum region 3
YL	lower boundary value used in an interval halving convergence procedure
YU	upper boundary value used in an interval halving convergence procedure
Y1	mole fraction of $\alpha$ MNT in the organic phase of the vessel currently being simulated
Y10	same as Y1 but for toluene
Y2	same as Y1 but for mMNT
Y3	same as Y1 but for $\alpha$ DNT
Y4	same as Y1 but for mDNT
Y5	same as Y1 but for a TNT

Y6	same as Y1 but for MTNT
¥7	same as Y1 but for TNBX
Y8	same as Y1 but for TNB
Y9	same as Y1 but for nitric acid
2A	diffusion coefficient for organic components in the organic phase
28	diffusion coefficient for organic components in the acid phase

### **Function BNTH**

The definitions of the following variables are identical to those in the main program: AKA, AKEQ, AKMA, AKMATL, AK10, AK12, AK3S, AK3W, AK4S, AK4W, AK5S, AK6S, AK7, AK8, CP(I), DH(I), E10, E21, E3S, E3W, E4S, E4W, E5S, E6S, E7, E8, FD, FMAX, RHQ1, RHQ10, RHQ11, RHQ12, RHQ13, RHQ14, RHQ2, RHQ3, RHQ4, RHQ5, RHQ6, RHQ7, RHQ8, RHQ9, RP, RR, SA, SB, SC, SD, SE, SG, SH, SI, SJ, TR

## Remaining variables are defined as follows:

A (1)	moles/hr of component i in the acid phase of the stream for which the enthalpy is currently being calculated	
CP11	equivalent to CP (11)	
CP12	equivalent to CP (12)	
CP13	equivalent to CP (13)	
CP14	equivalent to CP (14)	
CL.8	equivalent to GP (9)	
DLT	temperature of the vessel (relative to 20°C) for which the enthalpy of a particular exiting stream is currently being calculated	

O	ENTH	enthalpy of the stream currently being calculated (kcal/hr)
	ENTHO	enthalpy of mixing of the acid components in the acid phase of the stream currently being calculated based on the zero nitric curve of the McKinley and Brown data (kcal/lb)
	ENTH2	same as ENTHQ but based on the 20% nitric curve
	r	adjusted weight fraction (nitrobody free basis) of nitric plus equivalent sulfuric acid in the acid phase of the stream currently being calculated
	1	general index integer
	J	general index integer
	LBS	lb/hr of nitric acid, sulfuric acid, water and 80s flowing with the acid phase of the stream for which the enthalpy is currently being calculated
	LBSE	lb/hr of nitric acid plus equivalent sulfuric acid in the acid phase of the stream for which the enthalpy is currently being calculated
	P(I)	moles/hr of component i in the organic phase of the stream for which the enthalpy is currently being calculated
	T	moles/hr of a given organic component in the combined acid and organic phases (i.e., in the total stream) of the stream for which the enthalpy is currently being calculated
	TP	temperature of the vessel for which the enthalpy of a particular exiting stream is being calculated
	WTA	LBSE/LBS; nitrobody free weight fraction of nitric plus equivalent sulfuric acid in the acid phase of the stream for which the enthalpy is currently being calculated

是是一个时间,我们就是一个时间,我们就是一个时间,我们就是一个时间,我们就是一个时间,我们也是一个时间,我们也会一个时间,也是一个时间,也是一个时间,也是一个时间 1990年,我们就是一个时间,我们就是一个时间,我们就是一个时间,我们就是一个时间,我们就是一个时间,我们就是一个时间,我们就是一个时间,我们就是一个时间,我们

WTN	weight fraction of nitric acid in the nitric plus equivalent sulfuric acid portion of the acid phase of the stream for which the enthalpy is currently being calculated
X(I)	the ith data point on the zero % nitric curve from the enthalpy-concentration data of McKinley and Brown
Y (I)	the ith data point on the 20% nitric curve form the enthalpy-concentration data of McKinley and Brown

# APPENDIX I

Steady State Simulation--Input Data Format

CARD	DATA READ	FORMAT
1	AK12, AK3W, AK4W, AK58, AK68, AK7, AK6, AK10	8F10.0
2	AK38, AK48, E38, E48	8F10.0
3	E21, H3W, E4W, E5S, R6S, R7, E8, E10	8F10.0
4	SA, SB, SC, SE, SG, SH, SI	8F10.0
5	SJ, RR, TR, AKMA, AKMATL, FMAX FD, RP	8F10.0
6	AKA, AKS, AKC, AKEQ, AKEQOV, FOV	8F10.0
7, 8	AVI(I,1), I=1,14	8F10.0/6F10.0
9,10	PVI (I,1), I=1,14	8F10.0/6F10.0
11,12	AVI(I,2), I=1,14	8F10.0/6F10.0
13,14	PVI(I,2), I=1,14	8F10.0/8F10.0
15,18	AVI(I,3), I=1,14	8F10.0/6F10.0
17,18	PVI(I,3), I=1,14	8F10.0/8F10.0
19,20	AVI(I,4), I=1,14	8F10.0/6F .0
21,22	PVI(I,4), I=1,14	8F10,0/6F10.0
23,24	AVI(I,5), I=1,14	8F10.0/6F10.0
25,26	PVI(I,5), l=1,14	8F10.0/6F10.0
27,28	AVI(1,6), I=1,14	8F10.0/6F10.0
29,30	PVI(1,6), I=1,14	8F10.0/6F10.0
31,32	AVI(I,7), I=1,14	8F10.0/6F10.0
33,34	PVI(I,7), I=1,14	8F10.0/6F10.0
35,36	AVI(I,8), I=1,14	BF10.0/6F10.0
37,38	PVI (I,8), I=1,14	8F10.0/8F10.0
39,40	AVI(I,9), J=1,14	8F10.0/6F10.0
41,42	PVI(I,9), I=1,14	8F10.0/6F10.0
43,44	AVI (I,10), I=1,14	8F10.0/6F10.0

CARD	DATA READ	FORMAT
45,46	PVI(I,10), I=1,14	8F10.0/6F10.0
47,48	AVI(I,11), I=1,14	8F10.0/6F10.0
49,50	PVI(I,11), I=1,14	8F10.0/6F10.0
51,52	AVI(I,12), I=1,14	8F10.0/6F10.0
53,54	PVI(I,12), I=1,14	8F10.0/6F10.0
55,56	AVI(I,13), I=1,14	8F10.0/6F10.0
57,58	PVI (I, 13) , I=1,14	8F10.0/6F10.0
59,60	AVI(I,14), I=1,14	8F10.0/6F10.0
61,62	PVI(I,14), I=1,14	8F10.0/6F10.0
63	FACT 8, FACT 5, FACT 4, FACT 3, FACT 2	5F10.0
64	NDA	12
65	TITLE	20A4
86	IPRNT, EI, ILIST, IMBAL, IPI, IAFT, IPNCH, ICHECK	911
67	FIN(1,J), J=1,8	8F10.0
88	FIN(2,J), J=1,8	8F10.0
69	FIN(3,J), J=1,8	8F10.0
70	FIN(4,J), J=1,8	8F10.0
71	FIN(5,J), J-1,8	8F10.0
72	FIN(8,J), J=1,8	8F10.0
73	FIN (7, J), J=1,8	8F10.0
74-83	((CQMP(J,I), I=1,14), J=1,5)	8F10.0/6F10.0
84	NO(I), I=1,6	61
85	TEMP(I), I=1,8	8F10.0
86	VKM(I), I=1,8	8F10.0
87	VKM8	F10.0
88	EAQ(I), I=1,6	6F10.0
89	EØA(I), I=1,6	8F10.0

# APPENDIX J

Parameter Fitting

Table J1
Nitration snapshot
B-Line - 8/18/71 - 1:30 pm

Strong Nitric Flows, Ibs/hr	43 202 144	1118 	103.17 "0.95 15.42 0.57 -6.50 20.27
Weak Nitric Flows, Ibs/hr 500 from Fume Recovery 2445 from SAR	3467 from SAR	1200 lbs/ 5	103.28 90.53 15.15 2.50 -6.16 19.04
Weak Ni Flows, Il 500 from Recovery 2445 from	3467 fr	er to 1A -	100.86 88.32 13.97 4.33 -3.28 16.12
Recycle Setting	7 7 7 7 7 7 7 7 7 7 7 7 7 7 7 7 7 7 7 7	104 1 No fresh water added Yellow water to 1A - 1200 lbs/hr Acid Analysis - Acid Phase  1B 2 3B 4 5	98.26 84.93 13.86 6.58 -0.29 23.18
r inte	·	lysis - 2	88.49 78.65 10.60 4.11 9.81 7.12
Separator Temperature 51	51 66 78 94 103	104 Acid Anz 1B	72.78 66.67 5.89 1.96 26.54 0.49
		ır ır Spent	74.68 67.22 3.63 12.00 23.41 0.38
Nitrator Temperature 51	44 76 86 89 89 96	2418 lbs/hr 12300 lbs/hr Yellow Water	11.97 8.28 4.76 86.98
<b>1</b> A	1B 2 3A 3B 4	6 Toluene Oleum	Total Acidity Total Sulfuric Actual Nitric HNOSO4 Water Nitrobody Acid Density

Table J1 (Cont'd)

		Acid Ana	dysis - Ni	Acid Analysis - Nitrobody Phase	hase		
		118	2	38	4	ro.	ဖွ
Total Acidity		4.88	9.37	4.88	1.06	0.71	0.16
Actes Sulfuric		0.70	1.51	1.54	0.63	0.60	0.12
MCIUAL NITTO		4.97	96.6	3.96	0.28	0.14	0 05
Motor and with	,	0.81	0.24	99.0	0.66	0.33	; ; ;
water and Nitropody	ody	99.42	88.33	93.99	98.93	99.17	60 60 60
Separator Density		1.208	1.366	1.430			
			Gas Analysis	Sis			
Gas	쥥	aphy – Per	rcentage by	r Volume	MSA Billionaire		Total Tolumo
	8	Z	8	NO	NOX	TININ	Loss
Sample 1 6.89 Sample 2 6.24	17.62 17.84	63.3 <b>8</b> 69.31	5.07	0.46	3.0	610	7.5
Total Gas Flow - 687 cu ft/min	687 cu ft/miy	8			3		r.
			Acid Phase	<b>8</b> {			
	Oxidatio	Oxidation Products (Wt \$)	(Wt \$)				
Compound	Spent						
	Acid	118	2	38		2	9
Trinitrobenzene	0.50	0.04	0.22	0.14	0.02	0.01	

Table 31 (Cont'd)

Phase	
Acid	
y in	
Nitrobod	

<b>6</b>				,	0.01	0.05	0.16	0.0		0.26	86.69	0.05	2.02	1.01	99.72
us					90.0	0.03	1.50		0.01	1.60	95.72	0.04	1.76	19.0	98.36
4					0.54	0.12	16.99	0.01	0.05	11.71	86.20	0.02	1.49	0.57	88.28
8					3.28	0.3	36.42	0.03	0.37	40.41	57.89	0.02	1.16	0.39	29.46
2					10.63	0.56	84.50		0.84	96.53	2.85		0.33	9 <b>6</b>	3.24
118	43.93	28.36	75.33		3.67	0.07	16.75			20.49	4.13				4.13
Spent Acid	25.67	19.04	46.65	2.06	5.72	0.19	33.03			38.94	11.84				11.84
Compound	2-MNT 3-MNT	4-MNT	TOTAL MAT	*PNM	2,6-DNT	2,5-DNT	2,4-DNT	3,5-DNT	3,4-DNT	TOTAL DNT	2,4,6-TNT	2,3,5-TNT	2,4,5-TNT	2,3,4-TNT	TOTAL TINT

\*PNM - phenylnitromethane resulting from methyl group nitration of toluene

Table J1 (Cont<sup>4</sup>d)

# Nitrobody Phase

_
<u>ت</u>
E
Products
<b>Exidation</b>

•	99.0		•						0.01	0.01	0.16	<b>5</b> .0		0.22	96.00	90.0	2.32	1.33	99.71	
w	0.19		rs.						0.10	9.6	1.47	0.001	0.01	1.62	39.76	0.02	2.29	1.22	98.20	
•	0.25		•						0.89	0.13	9.12	0.01	0.12	10.27	86.39	0.63	2.05	1.02	89.48	
8	0.37	Phase	8						4.13	0.13	25.35	0.02	0.55	30.36	66.92	0.02	1.64	69.0	69.27	1.430
2	0.30	Analysis of Nitrobody Phase	2						10.99	0.47	58.12		1.28	70.86	28.50		0.25	0.10	28.85	1.366
118	0.02	Analysis	118	0.28	45.98	2.87	28.26	77.11	2.31	0.12	15.65		0.12	18.20	4.23				4.23	1.208
Compound	Trinitrobenzene		Compound	Toluene	2-MNT	3-MNT	4-MINT	TOTAL MNT	2,6-DNT	2,5-DNT	2,4-DNT	3,5-DNT	3,4-DNT	TOTAL DNT	2,4,6-TNT	2,3,5-TNT	2,4,5-TNT	2,3,4-TNT	TOTAL TNT	Separator - Density

Table J2
Final values of parameters

Parameter	Coded Name	Value	Units
k <sub>12</sub>	AK12	88.9	dimensionless
k <sub>3s</sub>	AK38	1.8x10 <sup>†</sup>	hr <sup>-1</sup>
k <sub>48</sub>	AK48	1.0x10 <sup>7</sup>	hr <sup>-1</sup>
k <sub>55s</sub>	AK58	124,5	hr <sup>-1</sup>
k <sub>6s</sub>	AK68	506,8	hr <sup>-1</sup>
k <sub>7</sub>	AK7	.09	hr <sup>-1</sup>
k <sub>a</sub>	AKB	2,1	hr <sup>-1</sup>
k <sub>10</sub>	AK10	.001	hr <sup>-1</sup>
k <sub>a</sub>	AKA	.2	(ft <sup>8</sup> ) (lb-moles) -1
K <sub>eq1</sub>	<b>S</b> H	.03	(lb-moles) $(ft^8)^{-1}$
K <sub>eq2</sub>	AKEQ	50.	dimensionless
Keq3	SJ	6.0x10 <sup>4</sup>	$(lb-moles)^{3} (ft^{3})^{-2}$
K <sub>eq4</sub>	8C	0.	(ft <sup>2</sup> ) (lb-moles) -1
E <sub>21</sub>	B21	88.9	(Kcal) (lb-mole) -1
E <sub>3s</sub>	E3S	2.2x10 <sup>4</sup>	(Kcal) (lb-mole) -1
E <sub>4s</sub>	E48	1.0x10 <sup>4</sup>	(Kcal) (lb-mole) -1
E <sub>5s</sub>	E58	9.3×10 <sup>8</sup>	(Kcal) (lb-mole) -1
E <sub>6s</sub>	E6S	6.5×10 <sup>8</sup>	(Kcal) (lb-mole) -1
E <sub>7</sub>	<b>E</b> 7	5.0x4x10 <sup>8</sup>	(Kcal) (lb-mole) -1

Ü	Parameter	Coded Name	Value	Units
	E <sub>8</sub>	E8	7.0x10 <sup>8</sup>	(Kcal) (lb-mole) -1
	E <sub>10</sub>	E10	1.0	(Kcal) (lb-mole) -1
	k <sub>m</sub> a	AKMA	2.75x10 <sup>4</sup>	[(lb-moles)(hr) $_{-1}^{-1}$ (ft <sup>2</sup> ) $_{-1}^{-1}$ ] × [(ft <sup>2</sup> )(ft <sup>2</sup> )]
	(k <sub>m</sub> a) <sub>p</sub>	AKMAP	1.55×10 <sup>4</sup>	[(lb-moles) (hr) $^{-1}$ (ft <sup>2</sup> ) $^{-1}$ ] $<$ [(ft <sup>2</sup> ) (ft <sup>2</sup> ) $^{-1}$ ]
	(k <sub>m</sub> a) <sub>Tol</sub>	AKMATL	5.0x10 <sup>8</sup>	[(lb-mole)(hr) $^{-1}$ (ft <sup>2</sup> ) $^{-1}$ ] × [(ft <sup>2</sup> )(ft <sup>3</sup> )]
	į	SI	1.51	dimensionless
	F <sub>D</sub>	FD	. 65	dimensionless
	RP	RP	.6	dimensionless

APPENDIX K
Steady State Optimization

# Table K1

# Independent variables

Variable	Rank
Weak Nitric Acid to 1A	6
Weak Nitric Acid to 1B	20
Weak Nitric Acid to 2	5
Fume Recovery Nitric Acid to 1A	18
Fume Recovery Nitric Acid to 1B	19
Yellow Water to 1A	22
Yellow Water to 1B	28
Yellow Water to 2	17
Fresh Water to 1A	26
Fresh Water to 2	27
Strong Nitric Acid to 3A	4
Strong Nitric Acid to 3B	12
Strong Nitric Acid to 4	3
Strong Nitric Acid to 5	11
Strong Nitric Acid to 6	2
Oleum to 6	1
Concentration of Weak Nitric Acid	13
Concentration of Fume Recovery Nitric Acid	10
Concentration of Strong Nitric Acid	9
Sulfuric Content of Oleum	8
remperature of Nitrator 1A	7
Femperature of Nitrator 1B	25
Cemperature of Nitrator 2	21
Cemperature of Nitrator 3A	14
Cemperature of Nitrator 3B	18

# Table K1 (Cont'd)

Variable	Rank
Temperature of Nitrator 4	23
Temperature of Nitrator 5	24
Temperature of Nitrator 8	16
Recycle Valve Notch Setting in Separator 1	32
Recycle Valve Notch Setting in Separator 2	31
Recycle Valve Notch Setting in Separator 3	29
Recycle Valve Notch Setting in Separator 4	30
Recycle Valve Notch Setting in Separator 5	33
Recycle Valve Notch Setting in Separator 6	94

### Table K2

### Constraint list

- 1. § DNT in Crude TNT must be less than .4
- 2. § Dissolved Nitrobody in Spent Acid  $\leq .5$
- 3. § DNT in Dissolved Nitrobody  $\leq 55$ .
- 4. Weak Nitric Concentration ≤ 61%
- 5. Fume Recovery Concentration ≤ 58%
- 6. Strong Nitric Concentration ≤ 99%
- 7. % H<sub>2</sub> SO<sub>4</sub> in Oleum > 57%
- 8. Wt § Actual Nitric (AN) in stages 3-6  $\leq$  16
- 9.  $AN(3) \ge 7$
- 10.  $AN(4) \ge 8$
- 11.  $AN(5,6) \ge 10$
- 12.  $AN(1) \le 6$
- 13.  $AN(2) \le 11$
- 14. AN(I)  $\geq 2.5$
- 15. AN(2)  $\geq$  4.5
- 16. 20. < [Temperature of Nitrator 1A (T1A)]
- 17. 20. ≤ TIB ≤ 60. °C
- 18. 50 ≤ T2 ≤ 70. °C
- 19. 60. ≤ T\$A ≤ 100, °C
- 20. 60.≤ T3B ≤ 100. °C
- 21. 70.≤ T4 ≤ 110. °C
- 22. 80.≤ T5 ≤ 115. °C
- 23. 90,≤ T8 ≤ 120. °C
- 24. Heat Load in Nitrator  $1A \le 2.5 \times 10^6$  BTU/hr
- 25. 66.  $\leq$  (% Total Acidity of Spent Acid)  $\leq$  72.

Table K3

## Optimum operating conditions

Basis: 59.2 tons/day production rate 10000 lb/hr oleum feed rate

Nitrator	1A	1 <b>B</b>	2	3A	3B	4	5	6
Week Nitric	2000		1900					
Fume Recovery Nitric	600							
Yellow Water	1000							
Strong Nitric				600	500	400	100	1100
Oleum								10000
Toluene	1763	587						
Temperature	51	33	84	83	85	90	95	110

Stage	Actual Nitric	Total Acidity
1	3.1	70.9
2	6.1	85.0
3	11,1	93.6
4	13.5	101.2
5	13.6	103.6
6	13.9	104.2

Yield = 89.2%

DNT in crude TNT = .17%

Wt % dissolved nitrobody in spent acid = .3

Wt % of dissolved nitrobody which is DNT = 19.7

Raw Material Cost = 5.45\*/lb TNT

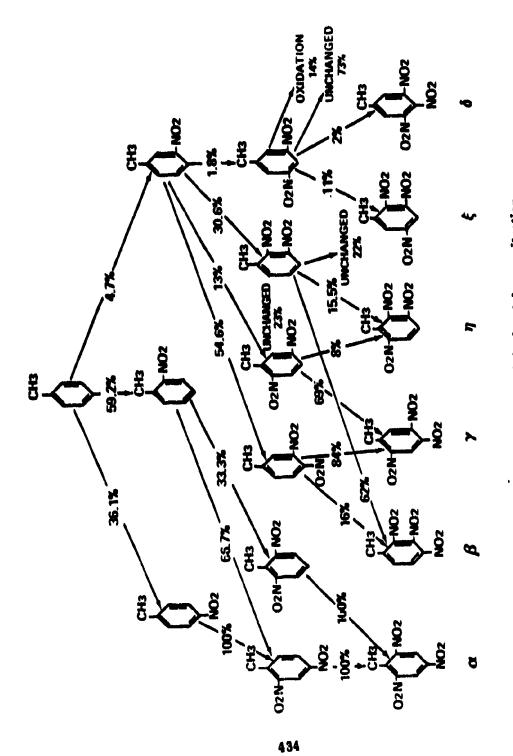


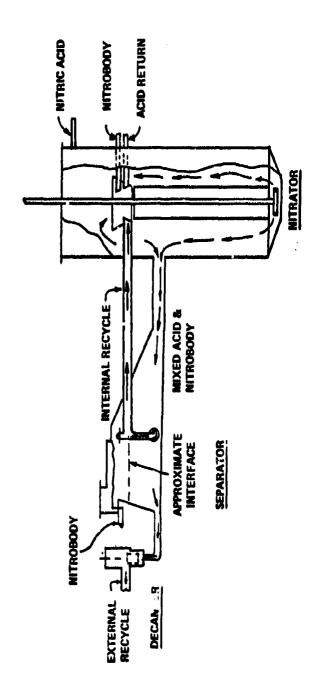
Fig 1 Isomers formed during inluene nitration

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Fig 2 Oxidation of trinitrotoluene

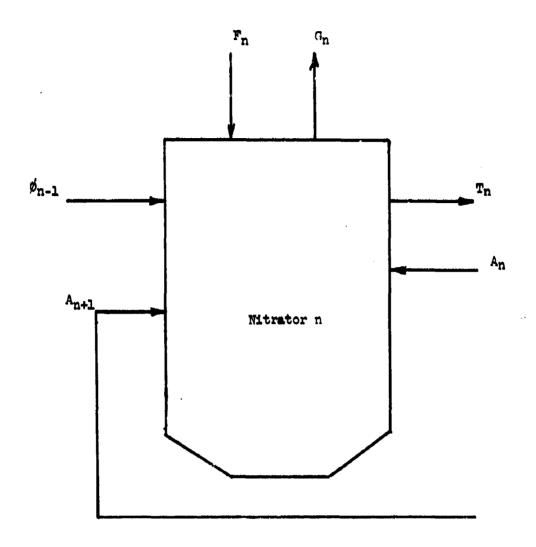
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Fig 3 TNT nitration flow scheme (inter-vessel relationship)



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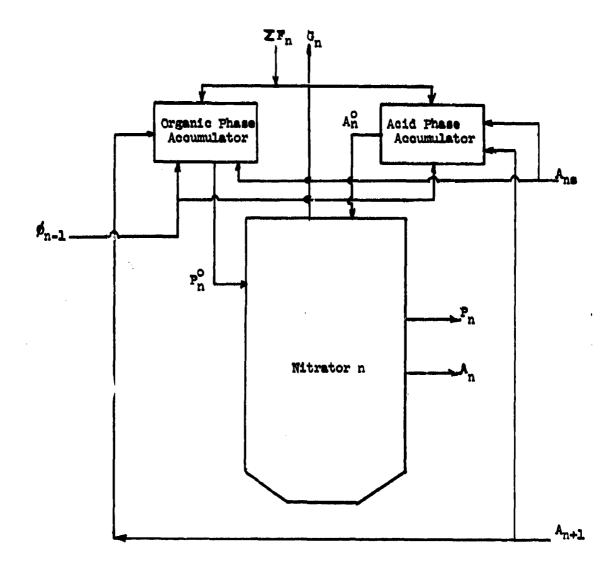
Fig 4 Nitrator-separator combination



where:

 $\Sigma$ F<sub>n</sub>\* sum of fresh feeds to nitrator n  $\phi_{n-1}$ = organic stream forward from separator n-l  $G_n$ \* total off-gas  $A_{n+1}$ = acid recycle stream from separator n+l  $A_n$ = acid recycle stream from separator n  $T_n$ = total outflow from nitrator n

Fig 5 Generalized nitrator



### where:

 $P_n^0$  = total organic phase input to nitrator n

 $A_n^0$  = total acid phase input to nitrator n

 $P_n$  = total organic phase output from nitrator n  $A_n$  = total acid phase output from nitrator n

 $A_{ns}$  is equivalent to  $A_n$  in figure 5 and  $A_{n+1}$ ,  $\phi_{n-1}$ ,  $G_n$  and  $\sum F_n$  are defined exactly as in figure 5

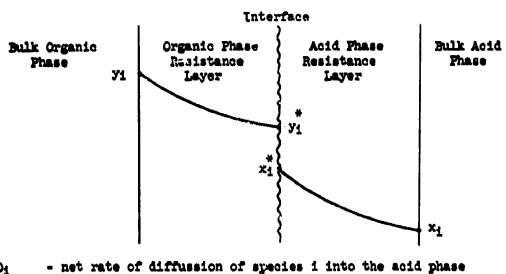
Fig 6 Simplified nitrator

CHEMICAL SPECIES CONSIDERED

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GAS PHASE	XON	8 *	Tree											
ACID PHASE	TOLUENE	CHANT	T NJES	TMO»	MDNT	*TNT	MTMT	ENT.	TNBX	HNO <sub>3</sub>	H <sub>2</sub> SO4	H <sub>2</sub> 0	803	HNOSO
ORGANIC PHASE	TOLUENE	TNM ->	M - MNT	≪ DNT	M - DNT	≪-TNT	M - TNT	TNB	TMBX	HNO <sub>3</sub>				

Fig 7 Cherrical components considered in the model



 $D_1$ 

- mole fraction species i in bulk organic phase Уí

- mole fraction species i in bulk acid phase Xi

- equilibrium solubility of all organic species in the acid phase or mole fraction organic species in acid phase at saturation xAq

- mass transfer coefficient in acid phase mp

- mass transfer coefficient in organic phase  $oldsymbol{\eta}_\mathtt{A}$ 

- moles species i in acid phase A<sub>4</sub>

ø, - moles species i in organic phase

- total moles in acid phase  $A_{T}$ 

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**A**T - total moles in organic phase

- total moles of nitrobody in acid phase ANE

- total moles of nitrobody in organic phase **PNB** 

- superscript denoting equilibrium at the interface

Fig 8 Diffusion in the nitration process

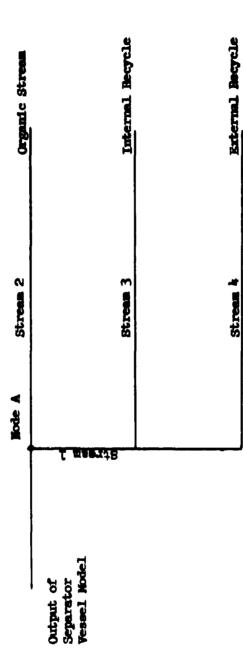
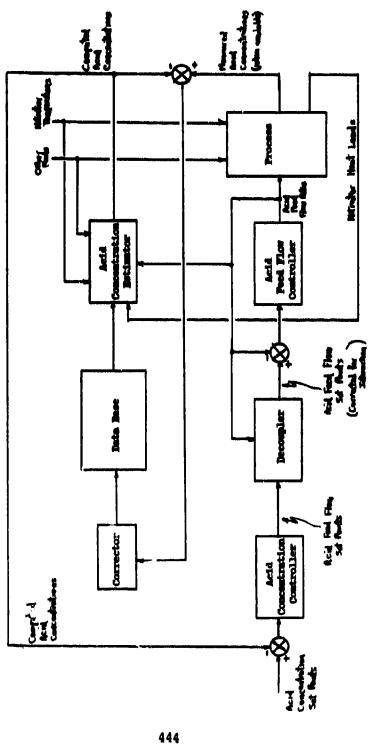


Fig 9 The separation process

# LIST OF VARIABLES

	Manipulated Variable	Controlled Variable
1.	Water to Stage 1	Total Acidity, Stage 1
2.	Weak Nitric to Stage 1	Nitric Concentration, Stage 1
3.	Water to Stage 2	Total Acidity, Stage 2
4.	Weak Nitric to Stage 2	Nitric Concentration, Stage 2
5.	Strong Nitric to Stage 3A	Nitric to Sulfuric Ratio, Nitrator 3A
6.	Strong Nitric to Stage 3B	Nitric to Sulfuric Ratio, Stage 3B
7.	Strong Nitric to Stage 4	Nitric to Sulfuric Ratio, Stage 4
8.	Strong Nitric to Stage 5	Nitric to Sulfuric Ratio, Stage 5
9.	Strong Nitric to Stage 6	Nitric to Sulfuric Ratio, Stage 6

Fig 10 Acid concentration control--variables operated on



Acid concentration control -- schenatic diagram Fig 11

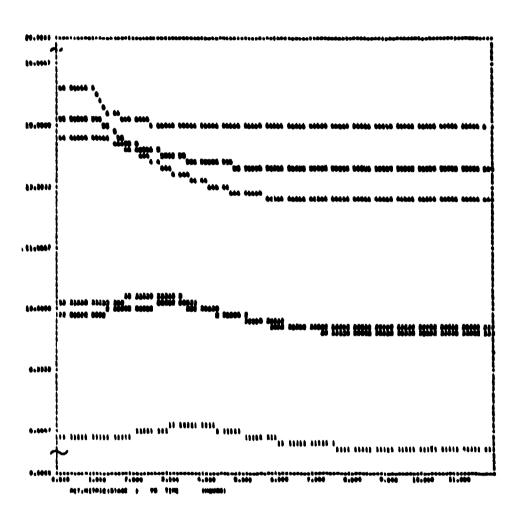
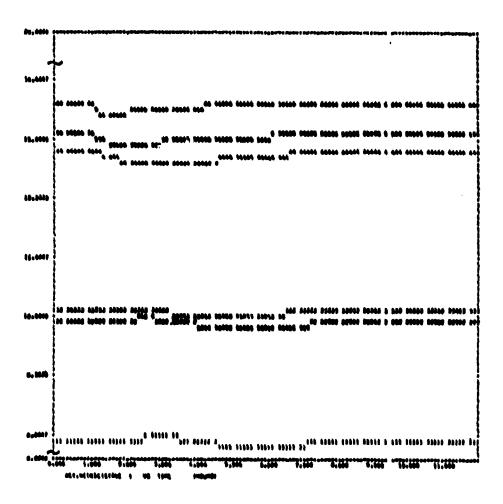


Fig 12 Open loop response of dynamic simulation



rig 13 Response of dynamic simulation under acid concentration control